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GREAT LAKES RESEARCH DIVISION

Technical Report

NONDESTRUCTIVE ACTIVATION ANALYSIS OF ENVIRONMENTAL SAMPLES

Richard Dams
John A. Robbins

John W. Winchester
Project Director

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I. Neutron Activation Analysis.

Introduction

The qualitative and quantitative determination of minute quantities by a technique known as activation analysis has become increasingly popular and is now generally recognized as a useful technique in various areas of scientific investigation and in an ever growing number of industrial control applications.

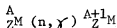
In this technique a nonradioactive substance is made radioactive by particle bombardment, and its decay characteristics are then measured. The nature of these characteristics allow an identification and the magnitude is a measure of the concentration. The sensitivity is extremely high for a large number of elements. The specificity is usually excellent. A number of textbooks describing the principles, the possibilities and a number of applications are available (1-5).

1. Principles.

Activation analysis is the determination of the weight of a particular element in a sample by measuring the radioactivity induced by irradiation, usually with thermal neutrons, but in some cases with fast neutrons or even with photons or charged particles.

The most commonly used and by far most intense neutron sources are provided by neutron chain reactors utilizing the fission reaction. Fast neutrons (>1 Mev) produced by the fission of uranium are moderated to epithermal (~ 0.4 ev) and further to thermal (<0.4 ev) energies. These thermal neutrons assure continuation of the fission reaction but can also be used to produce artificial radioactivity induced when a target material is exposed to thermal energies a chance exists that another nuclide will be created by neutron capture, immediately followed by emission of electromagnetic radiation.

The reaction is as follows

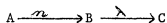


Examples are ${}_{11}^{23}\text{Na} (n, \gamma) {}_{11}^{24}\text{Na}$
 ${}_{27}^{59}\text{Co} (n, \gamma) {}_{27}^{60}\text{Co}$

The nuclides formed, ${}_{Z}^{A+1}\text{M}$, ${}_{11}^{24}\text{Na}$, ${}_{27}^{60}\text{Co}$ are often unstable and disintegrate, usually by emission of beta rays to an excited state of another isotope which deexcitates immediately by the emission of gamma rays. Beta rays are electrons or positrons. The beta rays from a given radioactive species are emitted with a continuous energy distribution extending from zero up to a maximum value. The gamma rays emitted by an excited state of a nuclide consist of electromagnetic radiations and have well defined energies $E = h\nu$ (ν is the frequency of the electromagnetic radiation). Gamma and beta rays can be detected with special counting equipment. More details about decay mode and interaction of radiation with matter can be found in the excellent books by Friedlander et al. "Nuclear and Radiochemistry" (6), and B. Harvey, "Introduction to Nuclear Physics and Chemistry" (7).

2. Formation and decay rate of radioactive species.

A target material A exposed to neutrons leads to the formation of nuclei B, which disintegrate to produce C as follows:



The rate of disintegration per atom of B is a constant, λ , specific for nucleus B. Indeed within a fixed time (half-life = $t_{1/2}$) half of the radioactive nuclides B will disintegrate into C. Thus $\lambda = \ln 2 / t_{1/2} \text{ (sec}^{-1}\text{)}$. The rate of formation of B depends on: (1) the number of atoms A present, namely N_A ; (2) the neutron flux ϕ , the number of neutrons per second passing through an area of 1 square centimeter (i.e. $\phi \text{ cm}^{-2}\text{-sec}^{-1}$); (3) the activation cross section of A, namely σ , expressed in barns (1 barn = 10^{-24} cm^2).

The rate of accumulation of B is given by the rate of formation minus the rate of disintegration.

Thus: $\frac{dN_B}{dt} = \sigma \phi N_A - \lambda N_B$ (1)

After integration assuming that before irradiation $N_B=0$, one finds that at the end of irradiation t_1 , the number of atoms of B present equals:

$$N_B^{t_1} = \sigma \phi N_A \lambda [1 - \exp(-\lambda t_1)] \quad (2)$$

The number of disintegrations per second, or the radioactivity at the end of irradiation t_1 equals:

$$\text{Act}_B^{t_1} = N_B^{t_1} \lambda = \sigma \phi N_A [1 - \exp(-\lambda t_1)] = \sigma \phi N_A S \quad (3)$$

where $S = 1 - \exp(-\lambda t_1)$ is called the saturation factor. The radioactivity present at time t_d after the end of irradiation will be smaller and is given as:

$$\text{Act}_B^{t_d} = \text{Act}_B^{t_1} \exp(-\lambda t_d) \quad (4)$$

Thus knowing σ , λ or $t_{1/2}$, ϕ and N_A the radioactivity for each irradiation time and cooling time can easily be calculated. Approximate values for σ and $t_{1/2}$ are tabulated (6, 8). The neutron flux can be determined and is usually approximately known for each irradiation site. Neutron fluxes vary between 10^8 $\text{n cm}^{-2} \text{ sec}^{-1}$ up to several times $10^{14} \text{ n cm}^{-2} \text{ sec}^{-1}$ depending on the power of the nuclear reactor and on the irradiation site in the reactor. The number of atoms is given as

$$N_A = \frac{0.602 \times 10^{24} \times g \times \theta}{\text{M.W.}} \quad (5)$$

where g is the weight in grams
M.W. is the molecular weight of the target material
 θ is the isotopic abundance

3. Activation Analysis.

Inversely one obtains the weight of the element from the activity measurement by substituting equation (5) in (3)

$$g = \frac{\text{Act}_B^{t_1} \times \text{M.W.}}{0.602 \times 10^{24} \times \sigma \times \phi \times S \times \theta} \quad (6)$$

This technique is called absolute activation analysis. The values for σ are however not known accurately, a precise determination of ϕ is time consuming and an absolute measurement of the radioactivity is difficult. Therefore, a comparator method is usually used in activation analysis. A standard containing a well known

amount of the element is irradiated simultaneously with and as near as possible to the unknown sample. A relative counting of both samples is performed and the unknown weight is calculated as follows:

$$(g)_v / (g)_S = (Act)_v / (Act)_S \quad (7)$$

where $(g)_v$ and $(g)_S$ are the weights of unknown and standard $(Act)_v$ and $(Act)_S$ are countrates of unknown and standard.

Many sources of errors can be eliminated by irradiating the sample with a standard of similar composition. When the composition of a substance is unknown, a preliminary irradiation has to be performed and can be used as a qualitative analysis in order to fabricate a suitable standard. Care should be taken that the sample and standard are approximately the same weight, shape and thickness.

4. Sensitivity-Standard Deviations.

Neutron activation analysis is inherently a very sensitive technique for determination of a large number of elements. In the analytical and radiochemical literature several mathematical expressions are used as definitions for the limit of detection, ranging from one to ten times the standard deviation of the background and employing a widely varying terminology. Thus in order to draw valid conclusions with respect to detection capabilities from published data, it is necessary to examine carefully the definition and confidence level accepted by the author.

As can be seen from equation (6) the sensitivity of the analysis of an element by neutron activation is determined by its cross section (σ), its isotopic abundance (θ), the half-life of the isotope produced (λ), the irradiation time (t_i), and the neutron flux (ϕ). Furthermore, the sensitivity is determined by the efficiency of the detector and its background signal.

The following example shows how the sensitivity for an element can be calculated from equation (6).

Assume the following realistic values: $\sigma = 1$ barn

$$\theta = 1 \text{ (100\%)}$$

$$M.W. = 100$$

$$t_1 = t \text{ 1/2 thus } S = 0.5$$

$$\phi = 10^{12} \text{ n/cm}^2\text{-sec}$$

If the count rate necessary is 1 count per second with a detection efficiency of 33 percent the sensitivity for such an element is given as:

$$g = \frac{3 \times 100}{0.6 \times 10^{24} \times 2 \times 10^{12} \times 0.5 \times 1 \times 10^{-24}} = 10^{-9} \text{ grams}$$

If gamma counting is performed the abundance of the gamma rays in the decay scheme must be included.

Jenkins and Smales (9) calculated sensitivities assuming an irradiation at $10^{12} \text{ n/cm}^2\text{-sec}^{-1}$ to saturation or up to one month (which ever is shorter) and counting 2 hours after irradiation to allow for chemical separation. The lower limits of precise measurements were taken to be 100 dis/min, for beta emitters and 100 photons/min for gamma emitters. Table 1 summarizes the results.

As a matter of fact practical sensitivities may be much less favorable if a number of elements are determined simultaneously because irradiation cooling and counting time cannot be ideal for each isotope. An appropriate compromise must then be applied.

Radioactive disintegration being a statistical phenomenon the standard deviation of a counting equals the square root of the number of counts. If the net number of counts N is obtained by subtracting the background B from the total number of counts T. The standard deviation is calculated as follows:

$$\sigma_N = \sqrt{\frac{2}{T} + \frac{2}{B}} = \sqrt{T + B}$$

5. Counting-Gamma Spectrometry.

When a gamma ray strikes a scintillation counter (NaI(Tl)) it produces light flashes which release photoelectrons from a photosensitive electrode.

Element	Estimated sensitivity, grams
Eu, Au, Ho,	10^{-12}
As, Ir, In, Kr, La, Mn, Pr, Re, Sm, Se, Tb, Tm, W, Yb	10^{-11}
Sb, Ar, Br, Cs, Co, Cu, Er, Gd, Ga, Hf, Nd, Pd, P, Rb, Na, Ta, Th, U, Y	10^{-10}
Ba, Cd, Ce, Cl, Ge, I, Hg, Os, Pt, K, Ru, Ag, Te, Sn, Xe, Zn	10^{-9}
Bi, Mo, Ni, Se, Si, S, Tl,	10^{-8}
Ca, Cr, Fe, O, Sr, Zr,	10^{-7}
Pb	10^{-6}

Table 1. Sensitivity of activation analysis as calculated by Jenkins and Smales (9).

These are multiplied in a photomultiplier to produce a pulse which then can be further amplified and recorded.

When a gamma ray strikes a semiconductor diode (Ge(Li)) a number of electrons are lifted from the valence band to the conduction band and swept through the material under the influence of an applied electric field so that an electric pulse is obtained. This pulse can also be amplified and recorded.

Whenever the output from a detector-amplifier system is proportional to the energy dissipation in the detector the measurements of pulse heights may be very useful. Pulse height analysis is made versatile and rapid by the use of a very important tool in activation analysis, namely the multichannel analyzer. In the analogue to digital converter (A.D.C.) the pulses are sorted according to size and immediately recorded as counts in the appropriate channel of the memory unit. The resultant plot of the number of counts as a function of channel number is called a "gamma spectrum". The number of channels of an analyzer is determined by the capacity of the analyzer memory. Pulse height analysers with 50 up to 8192 channels are in use. The time required for sorting and storing a pulse is appreciable and the analyzer cannot process a new pulse during this "dead time".

Provision is usually made to lengthen the counting time because of these dead time losses. A multichannel should also include the ability to display the gamma-spectrum on an oscilloscope and to drive an automatic printer, plotter or magnetic tape unit so that the content of each channel can be appropriately recorded.

Gamma rays emitted by radionuclides being monoenergetic, it is possible to detect several isotopes in presence of each other if a response proportional to the original gamma-ray energy can be obtained from a detection unit. Thus if a scintillation crystal or a solid state detector is large enough to contain the entire path of the gamma ray, it can measure its total energy. This effect is called "photoelectric effect" and the resulting pulses corresponding to the total gamma energy form the so-called "photopeak". In addition to this there will be a number of "Compton" effects. This is an incomplete recovery of the gamma energy by the crystal and results in the appearance of a continuous distribution of smaller energies corresponding to the total energy minus the energy which escaped the crystal. This is called the "Compton continuum." High energy gamma rays striking a detector material can give rise to pair production. This involves the production of a positron-electron pair (energy necessary is $2mc^2 = 1.02 \text{ Mev}$). The positron is not stable and by the reverse process, positron-electron "annihilation", the same amount of energy is emitted as photons, namely two gamma rays with an energy of 0.511 Mev. If one of these or both escape the detector a peak is recorded with an energy equal to the gamma ray minus 0.511 or 1.022 Mev. They are called double or single escape peaks.

The most important characteristic of the gamma spectrum is the full energy peak resulting from photoelectric absorption of the gamma ray. Thus the detection efficiency for a full energy peak and the ability to resolve it from another peak with a slightly different energy are of prime importance. The energy resolution is defined as the "full width at half maximum". NaI(Tl) detectors have high inherently full energy detection efficiencies and can be obtained in large sizes

but unfortunately the energy resolution is rather poor (7.5% of the 0.662 Mev gamma ray of Cs^{137}) and is energy dependent. The Ge(Li) detectors on the other hand have lower intrinsic full energy detection efficiency and cannot yet be obtained in sizes larger than 100 cm^3 . The detection efficiency of a 30 cm^3 Ge(Li) detector may be 7 times lower at 0.2 Mev and 25 times lower at 1.3 Mev than the efficiency of a 3"x3" NaI(Tl) detector, but the energy resolution is much better, especially for high energy gamma rays. If coupled to good electronic apparatus an energy resolution of 2.5 Kev at 1.3 Mev is now easily obtainable. The resolution is only slightly energy dependent. Thus the resolution at 0.2 Mev is 8 times and at 1.3 Mev 25 times better than obtainable with NaI(Tl).

More detailed information about gamma spectrometry can be found in the following works, "Gamma Spectrometry" by Heath, R.L. (10) and "Applied Gamma-ray Spectrometry" by C. Crouthamel, F. Adams, and R. Dams (11).

6. Nondestructive Activation Analysis.

The very high energy resolution obtainable in the Ge(Li) detectors allows the simultaneous specific measurement of the radiation emitted by several isotopes. Thus it becomes possible to distinguish and determine isotopes in complex mixtures without any chemical separation. The resultant instrumental or nondestructive activation analysis is attractive for different reasons.

- 1) The sample remains essentially unchanged, and can be used for subsequent investigations after the decay of the induced activity. It also allows inter-laboratory comparison of results on the same specimen.

- 2) Time is gained. This allows the analysis of large numbers of samples, which is very important in environmental research in order to draw valid conclusions.

- 3) The instrumental approach allows the development of completely automated systems using computer handling of data. The analysis can be performed by an essentially unskilled operator.

4) Isotopes with very short half-lives can be determined, whereas during chemical separations many short-lived isotopes decay essentially completely before the count is begun.

II. Procedure for Non-Destructive Neutron Activation Analysis of Environmental Samples.

Experimental

1. Apparatus used.

Nuclear Reactor: The irradiations are performed in the Ford Nuclear Reactor of the University of Michigan, operating at 2 Megawatts. The reactor is a swimming pool type, where the water acts as coolant, moderator and reflector. Short-lived isotopes are produced sufficiently by short irradiations (less than 30 minutes) in the pneumatic tube facility (neutron-flux 2×10^{12} n/cm²-sec). The transit time of the sample is approximately 3 seconds and the sample holder is a special nylon tube (Rabbit). Longer lived isotopes are produced by longer irradiations (more than 1 hour) in a facility (H-5) in the immediate neighborhood of the core (neutron-flux 1×10^{13} n/cm²-sec). The samples are packed in a polyethylene bottle and are lowered in the pool by means of aluminum wire. Holes drilled in the side of the bottle provide sufficient cooling of the contents.

Counting Equipment: The samples are counted by high resolution gamma-ray spectrometry. The gamma spectrometer consists of,

Detector: Nuclear Diodes - True coaxial germanium-lithium drifted diode. It has a capacity of 27.5 pf. The detector is positioned in a vacuum chamber at 12 mm distance from a 0.5 mm aluminum window. Its performances are: a full width at half maximum (F.W.H.M.) of 2.5 Kev for the gamma transition of Co⁶⁰ at 1332.3 Kev, and a detection efficiency for this photpeak of 4% relative to a 3"x3" NaI(Tl) scintillation detector. This results in a peak to Compton ratio of 18.

Preamplifier: Nuclear Diodes-Model 101 is fixed to the detector chamber.

Linear Amplifier: Tenelec Model 202 - an inverted unipolar pulse shape is used. It can be differentiated once or twice.

Multichannel Analyzer: Nuclear Data 2200 Series - 4096 channel analyzer consisting of: Master Control; Analog to Digital Converter; Data reduce/integrate; Read in/out display; Memory Scaler; Memory Drivers; Memory array; Preset Counter; Magnetic

Tape Read; Magnetic Tape Control

Oscilloscope: Hewlett-Packard

Printer: Monroe-Data/Log - Model MC-13-80. It prints 20 channels per second.

Magnetic Tape Recorder: Kennedy 1400R - Incremental Magnetic Recorder 200 BPI.

It is adapted for use with Nuclear Data 2200 Series.

Counting Room: The equipment is used in a temperature and humidity controlled room.

Computer: IBM 360/67, with 2 CPU's and other on-line accessories, at the University of Michigan Computing Center.

2. Identification of Isotopes-Qualitative Analysis.

High resolution gamma-ray spectrometry is very valuable for identification of gamma-emitting isotopes. Indeed gamma-ray energies can be determined very precisely if proper care is taken to avoid instabilities and deviation from linearity of the analysing and amplifying equipment. Comparison of the gamma-ray energies with a compilation of energies of radionuclides formed by neutron irradiation (12, 13) generally allows an immediate identification of the gamma emitting isotopes present without further investigation of other decay properties, or radiochemical behavior. Because the production reaction(s) of these isotopes in a reactor are also given in these compilations, the identity of the original target atom is usually very obvious.

The spectrometer can be energy calibrated using a set of radionuclides with precisely known gamma-ray energies. Table 2 lists a number of calibration lines. For most applications a gain setting of 1 Kev/channel is very appropriate. Figure 1 shows the deviation from this setting for the actually used gamma-ray spectrometer in an energy range from 55 up to 2800 Kev, a range which covers more than 95% the gamma rays emitted by reactor produced isotopes. By visual inspection of the oscilloscope display the location of sufficiently intense peaks can easily be estimated with an accuracy of better than 0.1 F.W.H.M., which is here equivalent to approximately 0.25 Kev.

<u>Isotope</u>	<u>Energy (Kev)</u>
²⁴¹ Am	59.568 ± 0.017
¹³¹ I	80.166 ± 0.009
¹⁵³ Gd	97.43 ± 0.02
¹⁵³ Gd	103.18 ± 0.02
¹⁷⁷ Lu	112.97
¹⁴¹ Ce	145.44 ± 0.05
¹³⁹ Ce	165.84 ± 0.03
¹⁷⁷ Lu	208.36
²⁰³ Hg	279.12 ± 0.05
¹³¹ I	364.47 ± 0.005
¹⁹⁸ Au	411.776 ± 0.01
Annihilation	511.006 ± 0.02
²⁰⁷ Bi	569.65 ± 0.10
¹³⁷ Cs	661.59 ± 0.07
⁹⁵ Nb	765.83 ± 0.07
⁵⁴ Mn	834.84 ± 0.07
⁸⁸ Y	898.01 ± 0.07
²⁰⁷ Bi	1063.82 ± 0.28
⁶⁰ Co	1173.13 ± 0.04
⁶⁰ Co	1332.39 ± 0.05
²⁴ Na	1368.40 ± 0.04
²⁰⁸ Tl(ThC'') (D.E.)	1592.3 ± 0.13
²⁴ Na(D.E.)	1731.6 ± 0.16
⁸⁸ Y	1836.1 ± 0.07
²⁴ Na(S.E.)	2242.6 ± 0.14
²⁰⁸ Tl(ThC')	2614.3 ± 0.09
²⁴ Na	2753.6 ± 0.12

Table 2. Energy Calibration Lines

From comparison with a calibration curve (Figure 1) an energy determination is immediately obtained with an accuracy of mostly better than 0.5 Kev. For more accurate energy determinations or for routine identification purposes a computer assisted qualitative analysis is performed. From a large number of calibration points a relation between gamma-ray energy and peak location is obtained and approximated by a nth degree polynomial. The degree of the polynomial and its

coefficients are calculated by a least squares fitting computer routine. Small differences in gain and zero threshold are corrected by recalibration with a high energy and a low energy gamma ray. The identification is then performed by comparing the experimental energies of the peaks of a complex gamma-ray spectrum with the gamma-ray energies of a library present in the memory of the computer. This library contains 2000 gamma-radiation energies of about 250 reactor produced isotopes (12). For the identification, also the half-life of the isotope is compared to the cooling time (time between end of irradiation and time of counting) of the sample.

Chapter 3 about computer handling gives more detailed information about this qualitative analysis.

3. Quantitative Analysis of Germanium Lithium Gamma-Ray Spectra.

The quantitative determination of the count rate due to a certain isotope from the gamma-ray spectrum involves the measurement of the area included under its full energy peak. This determination must not necessarily be quantitative. It is indeed sufficient to measure only a fraction of this area, provided that the same fraction of the peak area in the standard sample is summed. This can easily be carried out as follows: (see Figure 2). If A_0 is the count rate in the channel in which the highest number of counts is recorded, the area P under the peak is given as follows:

$$P = A_0 + \sum_{i=1}^n A_i + \sum_{i=-1}^{-n} A_i$$

However the observed peak will usually be interfered by a contribution due to the Compton continuum of more highly energetic full energy peaks. The area Q due to the Compton continuum under the peak can be approximated as follows:

$$Q = Q' + Q'' = \sum_{n+m}^{2n+m} A_i + \sum_{-(n+m)}^{-(2n+m+1)} A_i$$

The net peak area N is calculated as,

$$N = P - Q$$

and bears (within the standard deviation of the counts obtained in the channels) a constant relationship to the total area contained in the peak. The method can be optimized in terms of experimental conditions such as the calibration and the resolution of the spectrometer and the values chosen for n and m .

If the gain is set for 1 Kev/channel n is often taken equal to the F.W.H.M. of the peak. m is mostly chosen slightly higher. For the spectrometer used in this work the resolution varies from 2.2 to 3.0 Kev depending on the energy of the peak. During long counting times a small variation in gain, due to instability of the temperature in the counting room, is often experienced, resulting in an increased F.W.H.M. Therefore n can be chosen 2, 3 or 4 and for m a value of 4 is mostly appropriate. The choice of n , depends on the stability of the gain, the complexity of the spectrum, the counting statistics and the energy of the peak.

The precision of the determination of the peak area can be predicted if the peak is free of important interferences due to other peaks. The standard derivation is given as

$$\sigma_N^2 = \sigma_P^2 + \sigma_Q^2$$

$$\sigma_N = \sqrt{P + Q}$$

This method can however not always be applied:

1) Sometimes another photopeak is present immediately above or below the photopeak to be measured, interfering with the calculation of Q' or Q'' . The Compton contribution is then calculated at another energy region which is free of interferences.

2) If the photopeak itself is interfered by the presence of another photopeak it is advisable to determine the abundance of the nuclide from another photopeak. If this is not possible the interference may be reduced by decreasing n or the peak area may be calculated by doubling the area under the less interfered half of the photopeak. As a matter of fact the accuracy of such a determination is limited.

Because this nondestructive activation analysis procedure should be applied for large number of routine measurements, some sort of automatic data reduction is often necessary. Simple as these procedures for determination of count-rates under peaks may be, some sort of automation is sometimes desirable. Accuracy may be improved by the elimination of human errors in the transcription of data and in the operation of a desk calculator. However human judgment is to be maintained by incorporating into the calculation procedures suitable criteria for checking data.

A large number of methods and computer programs have been developed for the analysis of gamma-ray spectra obtained either with NaI(Tl) scintillation detectors or with Ge(Li) semiconductor detectors, but most of these programs involve thorough inspection and very complicated handling of data. Such a program is very expensive and thus less suitable for analysis of a very large number of quite similar samples. The next chapter describes the program for automatic data reduction applied in this work.

4. Irradiation-Counting Scheme.

High resolution gamma-ray spectrometry using germanium lithium drifted detectors allows the simultaneous measurement of a large number of isotopes. The ideal irradiation, cooling and counting times for the production and detection of an isotope are $t_{irr} \approx t_{1/2}$, $t_{cool} \leq t_{1/2}$ and $t_{count} \approx 2 t_{1/2}$. When a large number of elements have to be determined simultaneously by activation analysis, the irradiation-counting conditions can of course not be ideal for each isotope. Considering the elemental composition of typical environmental samples, the sensitivities for the determination of the elements present and the half-lives of the isotopes produced, a scheme, involving several irradiations and countings which allows the determination of a maximum number of elements was devised.

a) Analysis of elements giving rise to short-lived isotopes: Preliminary short neutron irradiations of some environmental samples and several countings soon after the end of irradiation showed the presence of several short-lived isotopes

such as ^{28}Al , ^{52}V , ^{49}Ca , ^{66}Cu , ^{37}S , ^{51}Ti , ^{38}Cl , ^{80}Br , ^{27}Mg , ^{56}Mn , $^{116\text{m}}\text{In}$, and ^{24}Na . Table 3 shows the half lives and most prominent gamma-rays, for the analysis of the elements giving rise to these isotopes. The following irradiation-counting scheme was developed. Considering the half-lives of the isotopes produced it appeared appropriate to perform only a short irradiation and to start counting soon after the end of irradiation. A five-minute irradiation performed in the pneumatic tube from which a fast return of the sample is ensured. For such short irradiation at a moderate neutron flux (2×10^{12} n/cm²-sec), the sample can easily be packed in a poethylene vial with a regular cap or with a snap top. Even small amounts of liquid can be irradiated under these conditions. The vial containing the sample is placed in a special irradiation container (rabbit) and the experimenter himself can transfer his sample to the irradiation site through a pneumatic conveyor. Automatic or manual return after a predetermined time can be applied. Because the sample holder also becomes radioactive the sample is transferred to another vial before counting. If the sample contains an appreciable amount of air it is advisable to ventilate it before counting because neutron irradiation of the argon present in the air (0.9%) gives rise to the production of the isotope ^{41}Ar ($t_{1/2} = 110$ min). For the same reason the irradiation container must be opened in a well ventilated hood. Three minutes after the end of irradiation a 400 second count is started. The bulk of the activity is then due to very short-lived isotopes such as ^{28}Al , ^{52}V , ^{49}Ca , ^{51}Ti , ^{66}Cu and ^{37}S . When these activities have died down to a lower level a second count is started for 1000 seconds at 15 minutes after the end of irradiation. This counting is favorable for the measurement of the isotopes ^{38}Cl , ^{27}Mg , ^{80}Br , $^{116\text{m}}\text{In}$, ^{56}Mn , and ^{24}Na . Typical gamma spectra obtained by counting an environmental sample after 3 minutes and 15 minutes cooling are shown in Figure 4-5. The fixed irradiation-counting scheme is also illustrated in Figure 3.

Element	Isotope Produced	Half-life	Most prominent γ rays Kev
Al	^{28}Al	2.30 min	<u>1778.9</u>
V	^{52}V	3.77 min	<u>1434.4</u>
Cu	^{66}Cu	5.1 min	<u>1039.0</u>
Mg	^{27}Mg	9.5 min	844.0; <u>1014.1</u> ;
Ca	^{49}Ca	8.8 min	<u>3083</u> ; 4071;
Na	^{24}Na	15.0 hr	<u>1368.4</u> ; <u>2753.6</u> ;
Mn	^{56}Mn	2.58 hr	<u>846.9</u> ; <u>1810.7</u> ; 2112.8; 2522; 2657; 2962; 3371;
In	$^{116\text{m}}\text{In}$	54.0 min	138.4; <u>417.0</u> ; 818.8; <u>1097.1</u> ; 1293.4
Cl	^{38}Cl	37.3 min	<u>1642.0</u> ; <u>2166.8</u> ;
Br	^{80}Br	18.0 min	<u>617.0</u> ; 640.3; 665.7; 704.3; 1256.7;
I	^{128}I	25.0 min	<u>442.7</u> ; 526.3; 743.5;
Ti	^{51}Ti	5.81 min	<u>320.0</u> ; 608.4; 928.5;
S	^{37}S	5.1 min	<u>3102.4</u>

Table 3. Nuclear properties of short-lived isotopes. The gamma-rays used in the determination are underlined.

Standards: In order to perform a quantitative analysis it is necessary to relate the count-rate of each isotope to the amount of the target element present. Therefore it is appropriate to prepare a number of standard samples containing well known amounts of each element. These amounts have to be balanced carefully to obtain a reasonable activity of all the respective isotopes measured.

Aqueous solutions were prepared from analytical grade salts of the elements to be determined. Table 4 shows the concentrations of the elements in these solutions. The solutions containing the metals consist of diluted nitric acid. The halogens on the other hand are present in diluted ammoniacal medium. From these solutions more diluted mixtures of the elements are prepared. Three mixtures respectively containing Al, V, Cu, Mg, Na, Ca, In, and Mn for solution 1; Cl, Br and I for solution 2; and a separate solution 3 for titanium and manganese because the titanium solution contains an unknown amount of chloride and because the large amount of magnesium present in solution 1 interferes with the determination of manganese. Because these three solutions are very diluted with respect to several

elements it is advisable to prepare new solutions each 3 months. Amounts ranging from 50 to 200 λ as shown in Table 4 of these three solutions are spotted on respectively 3 pieces of ashless filter paper (4x5 or 4x2.5 cm). Solutions 1 and 3 are carefully dried under an infrared lamp while spotting. Due to the volatility of the halogens, even in this ammoniacal medium, this is not allowed for solution 2. These standard samples can be used several times provided that sufficient time is elapsed between two irradiations to allow for decay of the isotopes measured. Because sulphur is very insensitive by activation analysis a known amount of $(\text{NH}_4)_2\text{SO}_4$ can be weighed and irradiated. Amounts ranging from 50 to 200 mg are appropriate.

The filter papers containing the standard solutions are then irradiated following the same scheme as the unknown samples. Because some of the elements spotted (Na, Cl, Br, Mn) are also present as impurities in the ashless filter paper, a blank paper has to be irradiated and the amounts found in this paper are summed with the known amounts to obtain the total standard.

Flux monitor: Standard and sample cannot be counted simultaneously thus also the irradiation cannot be performed simultaneously. This assumes that the reactor flux does not vary significantly between the irradiation of standard and sample at the irradiation site. Frequent modifications in the composition of the reactor core makes this assumption however uncertain over a long period of time. Also the reproducibility of the irradiation position is unknown. Therefore a flux monitor is irradiated together with each sample and standard and used to correct for possible flux variations. A small foil of titanium (1 mg) is used as flux monitor and is counted at 13 minutes after the end of the irradiation for 20 seconds. The activity due to the isotope ^{51}Ti ($t_{1/2} = 5.8 \text{ min}$) produced by (n, γ) reaction is calculated and acts as a measure of the flux. This flux monitor is sealed in a polyethylene jacket. Due to the short half life of the isotope ^{51}Ti , the same monitor can be used almost indefinitely. Indeed, if the rate of analysis does not

Solution Number	Element	Concentration $\mu\text{g/ml}$	Volume Spotted λ	Amount Irradiated μg
1	Al	10.0	200	2.00
1	V	1.028	200	0.2056
1	Cu	25.12 ⁵	200	5.025
1	Mg	500.5	200	100.1
1	Ca	499.0	200	99.8
1	Na	51.4	200	10.28
1	Mn	1.214	200	0.2428
1	In	0.076	200	0.0152
2	Cl	264.0	50	13.2
2	Br	12.7 ³	50	0.636
2	I	12.8 ³	50	0.641 ⁵
3	Mn	24.28	100	2.428
3	Ti	415.0	100	41.5
4	S	100 mg (NH ₄) ₂ SO ₄	weighed	24.2 mg S

Table 4. Standard solutions of determination of elements giving rise to short-lived isotopes.

exceed one analysis per 35 minutes more than 98% of the activity has decayed for the next measurement.

Calculation of the Concentrations: The net-peak areas are calculated from the spectra obtained from standards unknown samples and flux monitor. The net activities induced in the standards and samples are divided by the activity in the simultaneously irradiated titanium flux monitor to obtain a relative activity per unit of integrated flux. The weights of the element (added and ashless filter paper blank) as present in the standards are then divided by the relative activities per unit of flux obtained for the respective isotopes. This yields a conversion factor for each element. This conversion factor is a constant and can be used for several months if the counting is performed in the same geometry conditions and if the performance and settings of the counting equipment do not vary appreciably. By multiplying the relative activities per unit of flux obtained from an unknown sample with this conversion factor, the total weight of the element (sample and blank) present in the sample is immediately obtained. By applying the

fixed irradiation scheme no decay corrections are necessary because all the samples are counted at the same time after irradiation as the standards.

The following formula shows this calculation.

$$(m + b)_i = (m + b)_s \cdot \frac{F_s}{A_s} \cdot \frac{A_i}{F_i} = K \cdot \frac{A_i}{F_i}$$

where: $(m + b)_i$ = the weight of the element present in sample i (sample + blank)

$(m + b)_s$ = the weight of the element present in standard s (spotted + blank)

A_i = the peak area of the isotope in the sample i

A_s = the peak area of the isotope in the standard s

F_i = the peak area of the flux monitor irradiated with the sample i

F_s = the peak area of the flux monitor irradiated with the standard s

K = the constant conversion factor for the element

b) Analysis of Elements Giving Rise to Longer-Lived Isotopes: Because a long irradiation favors the production of longer lived isotopes, a sample was irradiated in the pool of the reactor at a high neutron flux (1.5×10^{13} n/cm²-sec) for several hours. At different times after the end of the irradiation gamma spectra of the sample were taken. Up to 1 hour after irradiation the spectrum mostly shows the very short-lived isotopes discussed in the previous section. Between 1 and 10 hours after irradiation the high activity due to ⁵⁶Mn and ²⁴Na greatly mask the presence of other isotopes. After 20 to 40 hours cooling time a number of photopeaks due to the following isotopes become visible:

⁸²Br, ⁴²K, ^{69m}Zn, ⁶⁴Cu, ¹²²Sb, ¹⁴⁰La, ¹⁵³Sm, ^{152m}Eu, ²⁴Na, and in some samples ⁷⁶As, ⁷²Ga, ¹⁸⁷W, ¹⁹⁸Au

Isotopes with half-lives longer than 10 days only become visible in the gamma-ray spectrum after the complete decay of the isotope ⁸²Br. Bromine is indeed usually abundant in environmental samples, it is very sensitive for activation analyses and the isotope ⁸²Br has a very complicated gamma-ray spectrum. Twenty to thirty days after the end of irradiation the following isotopes can be

detected, ^{51}Cr , ^{46}Sc , ^{60}Co , ^{59}Fe , ^{65}Zn , ^{75}Se , $^{110\text{m}}\text{Ag}$, ^{124}Sb , ^{141}Ce , ^{203}Hg and ^{233}Pa , and in some samples ^{58}Co .

For the quantitative analysis of the elements K, Zn, Br, Cu, Sb, La, Sm, Eu, Ga, W, Au, Cr, Sc, Co, Fe, Se, Ag, Ce, Hg, Th, and Ni a number of environmental samples (up to 15) and a standard containing well-known amounts of these elements are irradiated simultaneously for 2 or 5 hours at a neutron flux of $1.5 \times 10^{13} \text{ n/cm}^2\text{-sec}$ in the immediate neighborhood of the core of the reactor. The samples and the standard are heat sealed in polyethylene tubes. After irradiation the samples and standard are unpacked and transferred to the bottom of glass vials. After a cooling period of 20 to 30 hours the samples are counted for 2000 seconds for the measurement of the isotopes with half lives ranging from 8 to 50 hours. The isotopes with half-lives longer than 10 days are measured from a 4000 second count after 20 to 30 days cooling. Because the half-lives of the isotopes to be determined are sufficiently long as compared to the counting time a correction for the different decay times of the samples can easily be made. Table 5 shows the elements determined, the isotopes counted, their half lives and the energies of their gamma rays. Figures 7 and 8 show gamma spectra of both countings while the irradiation procedure is illustrated schematically in Figure 6.

Standards: Carefully weighed amounts of analytical grade salts of the elements are dissolved in dilute nitric acid. Well balanced amounts of these solutions are mixed to obtain reasonable activities of all the respective isotopes after irradiation. The solutions of gold contained some chlorine. Therefore it could not be mixed with the other elements (precipitation of silver and mercury). Also the addition of bromine and tungsten would cause evaporation (hydrobromic acid) or precipitation (tungstic acid H_2WO_4). Therefore these three elements are in a separate solution. The contents of these solutions and the concentrations of the elements are given in Table 6. Because the concentrations of several elements in these solutions are relatively low, new solutions should be prepared

Element	Isotope Produced	Half-life	Most Prominent γ -rays (Kev)
Na	^{24}Na	15.0 hr	<u>1368.4</u> ; <u>2753.6</u> ;
K	^{42}K	12.5 hr	<u>1524.7</u> ;
Zn	$^{69\text{m}}\text{Zn}$	13.8 hr	<u>438.7</u> ;
Cu	^{64}Cu	12.8 hr	β^+ (<u>511.0</u>);
Br	^{82}Br	35.9 hr	<u>554.3</u> ; <u>619.0</u> ; 698.3; <u>716.6</u> ; 827.8; <u>1043.9</u> ; 1317.2; 1474.7;
As	^{76}As	26.3 hr	559.2; <u>657.0</u> ; 767.5; <u>1228.8</u> ;
Ga	^{72}Ga	14.3 hr	601.1; 630.1; 786.4; 810.5; <u>834.1</u> ; <u>1800.1</u> ; <u>2201.4</u> ;
La	^{140}La	40.3 hr	<u>328.6</u> ; 486.8; 815.4; <u>1595.4</u> ;
Sm	^{153}Sm	47.1 hr	69.6; <u>103.2</u> ;
Eu	$^{152\text{m}}\text{Eu}$	9.35 hr	121.8; 344.2; 841.6; <u>963.5</u> ; 1315.0; 1388.9;
Sb	^{122}Sb	2.75 da	<u>504.0</u> ; <u>692.5</u> ; 1256.6;
W	^{187}W	24.0 hr	72.3; 134.3; 479.3; 551.4; 618.1; <u>685.7</u> ; 772.9;
Au	^{198}Au	2.70 da	<u>411.8</u> ;
Sc	^{46}Sc	83.9 da	<u>889.4</u> ; <u>1120.3</u> ;
Cr	^{51}Cr	27.8 da	<u>320.0</u> ;
Co	^{60}Co	5.24 yr	<u>1173.1</u> ; <u>1832.4</u> ;
Fe	^{59}Fe	45.1 da	<u>1098.6</u> ; <u>1291.5</u> ; 192.5; 143.0;
Ni	^{58}Co	71.3 da	<u>810.3</u> ; 863.5; β^+ (511.0)
Zn	^{65}Zn	245.0 da	<u>1115.4</u> ; β^+ (511.0);
Se	^{75}Se	121.0 da	96.7; 121.8; 136.0; <u>264.6</u> ; 279.6; <u>400.7</u> ;
Ag	$^{110\text{m}}\text{Ag}$	260.0 da	657.8; 706.4; 763.9; 884.5; <u>937.2</u> ; <u>1384.0</u> ;
Sb	^{124}Sb	60.9 da	<u>602.6</u> ; 645.7; 722.8; 1436.8; <u>1690.7</u> ; 2090.6
Ce	^{141}Ce	32.5 da	<u>145.4</u> ;
Hg	^{203}Hg	46.9 da	<u>279.1</u> ;
Th	^{233}Pa	27.0 da	<u>299.9</u> ; <u>311.8</u> ; 340.3;

Table 5. Nuclear properties of long-lived isotopes. The gamma rays used in the determinations are underlined.

detected, ^{51}Cr , ^{46}Sc , ^{60}Co , ^{59}Fe , ^{65}Zn , ^{75}Se , $^{110\text{m}}\text{Ag}$, ^{124}Sb , ^{141}Ce , ^{203}Hg and ^{233}Pa , and in some samples ^{58}Co .

For the quantitative analysis of the elements K, Zn, Br, Cu, Sb, La, Sm, Eu, Ga, W, Au, Cr, Sc, Co, Fe, Se, Ag, Ce, Hg, Th, and Ni a number of environmental samples (up to 15) and a standard containing well-known amounts of these elements are irradiated simultaneously for 2 or 5 hours at a neutron flux of $1.5 \times 10^{13} \text{ n/cm}^2\text{-sec}$ in the immediate neighborhood of the core of the reactor. The samples and the standard are heat sealed in polyethylene tubes. After irradiation the samples and standard are unpacked and transferred to the bottom of glass vials. After a cooling period of 20 to 30 hours the samples are counted for 2000 seconds for the measurement of the isotopes with half lives ranging from 8 to 50 hours. The isotopes with half-lives longer than 10 days are measured from a 4000 second count after 20 to 30 days cooling. Because the half-lives of the isotopes to be determined are sufficiently long as compared to the counting time a correction for the different decay times of the samples can easily be made. Table 5 shows the elements determined, the isotopes counted, their half lives and the energies of their gamma rays. Figures 7 and 8 show gamma spectra of both countings while the irradiation procedure is illustrated schematically in Figure 6.

Standards: Carefully weighed amounts of analytical grade salts of the elements are dissolved in dilute nitric acid. Well balanced amounts of these solutions are mixed to obtain reasonable activities of all the respective isotopes after irradiation. The solutions of gold contained some chlorine. Therefore it could not be mixed with the other elements (precipitation of silver and mercury). Also the addition of bromine and tungsten would cause evaporation (hydrobromic acid) or precipitation (tungstic acid H_2WO_4). Therefore these three elements are in a separate solution. The contents of these solutions and the concentrations of the elements are given in Table 6. Because the concentrations of several elements in these solutions are relatively low, new solutions should be prepared

- A_s = the peak area of the isotope in sample s
 λ = the decay constant of the element
 t_i = the cooling time of the sample i
 t_s = the cooling time of the standard s

5. Sensitivity-Detection Limits

In the irradiation counting system adopted a first detection limit is defined as the amount necessary to record at least 10 counts ($\sigma = 35\%$) under the photopeak or a number of counts equal to the background of the detector in that energy region. Practical detection limits are however mostly higher due to mutual interference of the isotopes. Indeed the sensitivity to measure a photopeak of an isotope is decreased when the peak is sitting on the Compton continuum of higher energetic gamma rays due to other isotopes. Thus a second detection limit may be defined which is defined as the amount of the element necessary to obtain at least a net photopeak activity equal to the standard deviation on its calculation. Thus if the net activity is given as $N = P - Q$, the detection limit is defined as $\sqrt{P - Q}$.

Column 5 on Tables 7 and 8 shows detection limits for typical air samples. As a matter of fact these values depend largely on the composition of the sample. For example a very high concentration of aluminum or chlorine may mask the presence of other short-lived isotopes. Large concentrations of sodium or bromine may interfere with the measurement of the isotopes with half-lives ranging from 8 to 50 hours. This results in an important decrease of the sensitivity.

The sensitivity of the analysis of an element in an environmental sample is further limited by the blanks. Indeed, abundant elements such as Cl, Br, Na, K, Zn, Fe, Mn, Al, Cu, Sb, Cr, etc. are liable to be present in the filter material, holder, or solvent in which the sample is collected.

In the present investigation an element is considered as being present if the net concentration exceeds twice the standard deviation of the blank value.

Element	t _{irr.} flux	t _{count}	t _{decay}	Detection limit, ng, for typical air filter samples
S	5 min $2 \times 10^{12} \text{ n/cm}^2\text{-sec}$	400 sec	3 min	25,000
Ca	"	"	"	1,000
Al	"	"	"	40
V	"	"	"	1
Cu	"	"	"	100
Ti	"	"	"	200
Br	"	1000 sec	15 min	20
In	"	"	"	0.2
Mn	"	"	"	3
Mg	"	"	"	3,000
Na	"	"	"	200
Cl	"	"	"	500
I	"	"	"	---

Table 7. Sensitivity of 13 elements by short irradiation in adopted irradiation-counting scheme.

Element	t _{irr} flux	t _{count}	t _{decay}	Detection limit, ng, for typical air filter samples
K	5 hour $1.5 \times 10^{13} \text{ n/cm}^2\text{-sec}$	2000 sec	20-30 hours	75
Zn	"	"	"	200
Cu	"	"	"	50
Br	"	"	"	25
As	"	"	"	40
Ga	"	"	"	10
La	"	"	"	2
Sm	"	"	"	0.05
Eu	"	"	"	0.1
Sb	"	"	"	30
W	"	"	"	5
Au	"	"	"	1
Sc	"	4000 sec	20-30 days	0.3
Cr	"	"	"	20
Co	"	"	"	2
Fe	"	"	"	1,500
Ni	"	"	"	1,500
Zn	"	"	"	100
Se	"	"	"	10
Ag	"	"	"	10
Sb	"	"	"	8
Ce	"	"	"	20
Hg	"	"	"	10
Th	"	"	"	3

Table 8. Sensitivity of 23 elements by long irradiations in adopted counting scheme.

This standard deviation is determined by the accuracy of the blank determination and by the homogeneity of the blank material.

6. Possible Interferences

Besides the thermal neutrons there are always some fast or fission neutrons present at each irradiation site of a reactor. These high energy neutrons induce threshold reactions after collisions with atoms. Such threshold reactions are (n,p), (n, α), and (n, 2n) reactions. Some determinations based upon an (n, γ) reaction induced by thermal neutrons may be interfered by such threshold reactions.

For example the determination of element ${}^A_Z M$ by reaction (1) is interfered by reactions (2), (3), and (4) on elements ${}^{A+1}_{Z+1} M$, ${}^{A+4}_{Z+2} M$, and ${}^{A+2}_Z A$.



To estimate the degree of interference it seemed appropriate to determine the ratio of the fast to thermal neutron flux.

Determination of Fission to Thermal Flux Ratio: Determination of the fission flux in a reactor site requires the measurement of the production rate of an isotope by a threshold reaction. Very often the reaction ${}^{32}_{16}S(n, p) {}^{32}_{15}P$ is used to determine the value of an undisturbed fission flux. Thus from an amount of sulphur irradiated during a time t_1 and counted at a time t_2 after irradiation the fission flux is calculated as follows:

$$\phi_f = \frac{D_{32S} \exp(+\lambda t_2)}{\sigma_{32S} N_{32S} [1 - \exp(-\lambda t_1)]}$$

where ϕ_f = the mean fission flux in n/cm²-sec.

D_{32S} = the absolute disintegration rate of ${}^{32}_{15}P$ produced by (n,p) reaction on sulphur

σ_{32S} = the mean fission cross section for the reaction ${}^{32}_{16}S(n, p) {}^{32}_{15}P$

N_{32S} = the number of atoms ${}^{32}_{16}S$ present

λ = the decay constant of the isotope ^{32}P

This requires however an absolute measurement of the ^{32}P disintegration rate.

The interference due to a threshold reaction does not depend on the absolute value of the fission flux but only on its ratio to the thermal flux. Thus it is sufficient to compare the count rate of ^{32}P produced by the threshold reaction to the count rate of ^{32}P produced by the thermal reaction $^{31}\text{P}(n, \gamma)^{32}\text{P}$. This requires now only a relative measurement of ^{32}P . Thus from a simultaneously irradiated and counted amount of phosphorus the thermal neutron flux is given as:

$$\phi_{\text{th}} = \frac{D_{31\text{P}} \exp(+\lambda t_2)}{\sigma_{32\text{P}} N_{32\text{P}} [1 - \exp(-\lambda t_1)]}$$

where $D_{31\text{P}}$ = the absolute disintegration rate of ^{32}P produced by (n, γ) reaction on phosphorus

$\sigma_{32\text{P}}$ = the thermal neutron cross section for the reaction $^{31}\text{P}(n, \gamma)^{32}\text{P}$

$N_{32\text{P}}$ = the number of atoms ^{31}P present

The ratio of both fluxes equals now

$$\frac{\phi_f}{\phi_{\text{th}}} = \frac{D_{32\text{S}} \sigma_{32\text{P}} N_{32\text{P}}}{D_{31\text{P}} \sigma_{32\text{S}} N_{32\text{S}}} = \frac{\text{Act}_{32\text{S}} \sigma_{32\text{P}} N_{32\text{P}}}{\text{Act}_{32\text{P}} \sigma_{32\text{S}} N_{32\text{S}}}$$

where $\text{Act}_{32\text{S}}$ = the relative count rate of ^{32}P produced by (n, p) reaction on sulphur

$\text{Act}_{32\text{P}}$ = the relative count rate of ^{32}P produced by (n, γ) reaction on phosphorus

Experimental: Simultaneously known amounts of analytical grade $(\text{NH}_4)_2\text{SO}_4$ and $(\text{NH}_4)_3\text{PO}_4$ were irradiated at both irradiation sites used (pneumatic tube and reactor pool). Seven days after irradiation the salts were dissolved in diluted nitric acid and adequate amounts were counted by liquid scintillation using the Čerenkov effect. ^{32}P is a pure beta emitter with a maximum beta energy of 1.71 Mev and a half life of 14.2 days. The interfering activity due to ^{35}S produced by (n, γ) reaction on ^{34}S is a pure beta emitter with a maximum beta energy of only 105 Kev and a half life of 85 days. This energy is too low to generate Čerenkov scintillation. To check the purity of the samples a decay curve analysis was performed for 3 months. For all the samples half-lives ranging from 14.0 to 14.4

days were found and no indication of the presence of shorter or longer lived isotopes was detected. The results shown in Table 9, are calculated using the following generally accepted values for the cross sections (14,15). $^{31}\text{P}(n, p) \quad ^{32}\text{P} \sigma = 0.19 \text{ barn}$
 $^{32}\text{S}(n, \gamma) \quad ^{32}\text{P} \sigma = 0.005 \text{ barn}$

Irradiation Site	Activity	Activity	Ratio Activity (P) Activity(S)	Neutron Flux Ratio Thermal/Fission
	mg P c/min	mg S c/min		
Pneum. Tube	206,480	8,823	23.40	7.6
Reactor Pool	4,060,200	295,354	13.75	4.5

Table 9. Determination of thermal to fission flux ratio.

Calculation of the interferences: Based upon the experimentally found ratio of the fission to thermal neutron-fluxes and on the mean neutron fission cross sections as compiled by Neuerht and Zyp correction factors for possible interferences were calculated (Table 10- column 5). This correction factor equals the weight fraction of the interfering element which has to be subtracted from the element determined. Thus the corrected amount of element M present is given as

$$M = M_E - I_E K_{M.I.}$$

where M_E = the amount of element M experimentally found

I_E = the amount of interfering element I experimentally found

$K_{M.I.}$ = the correction factor for interference of element I on the determination of element M

The concentration ratios of interfering to interfered elements which give rise to an interference of 1% were also calculated. These ratios may be compared to concentration ratios found in actual air samples (Table 10, columns 6 and 7). It is obvious that in these samples a large correction (>5%) can only be expected in the analysis of magnesium, which is interfered by a threshold reaction on aluminum. Therefore this low interference was also determined experimentally by irradiating a solution of pure $\text{Al}(\text{NO}_3)_3$. It appeared that 100 micrograms of aluminum

Element Determined	Interfering Element I	Reaction	Irradiation Site	Correction Factor $K_{M.I.}$	Concentration ratio resulting in 1% interference I/M	Actual Concentration ratios in air I/M
Na	Al	$^{27}\text{Al}(n, \alpha)^{24}\text{Na}$	Pneum Tube	0.00015	65	2-8
Na	Mg	$^{24}\text{Mg}(n, p)^{24}\text{Na}$	"	0.00023	43	1-10
Mg	Al	$^{27}\text{Al}(n, p)^{27}\text{Mg}$	"	0.15	0.067	0.5-5
Mg	Si	$^{30}\text{Si}(n, \alpha)^{27}\text{Mg}$	"	0.00002	490	1-10
Al	Si	$^{28}\text{Si}(n, p)^{28}\text{Al}$	"	0.002	4.9	0.2-4
Al	P	$^{31}\text{P}(n, \alpha)^{28}\text{Al}$	"	0.00055	18	
K	Ca	$^{42}\text{Ca}(n, p)^{42}\text{K}$	React Pool	0.0013	7.6	0.5-5
Sc	Ti	$^{46}\text{Ti}(n, p)^{46}\text{Sc}$	"	0.000008	1,250	150-1500
Cr	Fe	$^{54}\text{Fe}(n, \alpha)^{51}\text{Cr}$	"	0.000014	720	50-500
Mn	Fe	$^{56}\text{Fe}(n, p)^{56}\text{Mn}$	Pneum Tube	0.0000083	1,200	10-50
Mn	Co	$^{59}\text{Co}(n, \alpha)^{56}\text{Mn}$	"	0.0000014	7,200	0.005-0.05
V	Cr	$^{52}\text{Cr}(n, p)^{52}\text{V}$	"	0.00002	510	0.5-5

Table 10. Nuclear interferences due to threshold reactions.

generates a false amount of magnesium equal to 20 ± 8 microgram. This results in a correction factor $K_{\text{Mg-Al}} = 0.20 \pm 0.08$ which is in fairly good agreement with the calculated value of $K_{\text{Mg-Al}} = 0.15$. Magnesium concentrations are therefore corrected with this factor (0.20) in actual analysis.

7. Neutron Flux Mapping

In the pool of the reactor up to 15 samples can be irradiated simultaneously in a polyethylene bottle with a diameter of 4 cm and a height of 8.5 cm. For the calculation of the concentrations it is assumed that all the samples receive the same neutron flux as the standard sample. However in the immediate neighborhood of a finite reactor core an important vertical and horizontal neutron flux gradient may exist. The samples are thus liable to undergo a different flux.

Experiment: An experiment was devised to determine the importance of this effect at the irradiation site used. An aluminum wire was stretched along the walls of the polyethylene bottle. After irradiation it was cut into pieces of 1

to 2 cm. (see Figure 9). These pieces were carefully weighed, counted and the specific activities were related to the neutron flux at the respective sites. The ^{72}Ga activity produced by the reaction $^{71}\text{Ga}(n, \gamma)^{72}\text{Ga}$ on the gallium impurity was counted as a measure of the thermal neutron flux and the ^{24}Na activity produced by the threshold reaction $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ on the aluminum matrix was counted as a measure of the fission flux. These isotopes were counted by selecting their most prominent photopeaks respectively at 834 and 1368 Kev by means of a single channel analyzer coupled to a NaI detector. The result of one of these experiments is illustrated in Figure 9. As expected the fast neutron flux is more subject to gradients. Indeed a further thermalization sums up with the geometrical effect. The total horizontal gradient (4 cm) appears to be between 6 to 10% for the thermal and 25 to 35% for the fast fission flux. The total vertical gradient (8 cm) on the other hand is between 16 and 21% for the thermal and fission flux.

All the elements, except one (Ni) are determined from mainly thermal neutron produced isotopes. However also the thermal gradients may cause non-neglectible errors. Therefore, only the lower 4 cm of the bottle are filled with one row of vertically stacked samples, with the standard in the middle. This largely reduces errors due to vertical gradients. Errors due to horizontal gradients are minimized by turning the bottle over approximately 180° at half of the irradiation time.

III. Computerized Data Reduction

1. Introduction

From the foregoing discussion it can be seen that complete analysis of Ge(Li) gamma spectra involves the steps indicated in Fig. 10. Data from the storage volume must be transmitted to the appropriate processing device. In the case of hand analysis this would involve a paper tape listing of the spectrum channel by channel or an oscilloscope display. Paper tape printout of 4096 channels takes minutes even with high speed printout devices, while writing the data on magnetic tape for later processing requires about 30 seconds or less. For computer processing, spectral data stored magnetic tape must be read into fast memory core storage.

Next, all statistically significant peaks in the spectrum must be located. Doublets should be recognized and Compton edges should be discriminated against. While this process of peak recognition may be easily accomplished by eye, it is a non-trivial problem for computer. Various computer techniques have been devised such as the second difference method of Mariscotti,⁽¹⁶⁾ cross-correlation analysis of Black,⁽¹⁷⁾ the tangent method of Gunnink⁽¹⁸⁾ and others. In general, while all methods are successful in locating large isolated peaks, they may fail to work reliably in resolving doublets, discriminating between real peaks and Compton edges and in identifying small peaks which are marginally above the Compton continuum baseline.

A problem of comparable difficulty is that of determining net peak areas. Analysis of peaks which are large in comparison with a relatively flat baseline can be accomplished by summation over a specified interval centered on the peak maximum with subtraction of baseline counts estimated from channels on each side of the peak as mentioned in Chapter 2. In the case of peaks located near Compton edges where the continuum changes rapidly from channel to channel, this procedure can still be used successfully provided a judicious selection of channels is made for the estimate of the continuum under the peak. Difficulties arise in programming

computers to make the appropriate choices. Ralston and Wilcox⁽¹⁹⁾ have developed a procedure for calculating the baseline using an iterative spectrum smoothing routine discussed below. This baseline spectrum is probably a better approximation generally to the Compton continuum than the so-called straight line approach above but is limited in its ability to handle cases of broad peaks located near Compton edges. Additional problems arise in estimating **net** peak areas from resolved doublets. If the peaks are of comparable height, areas of individual components may be proportioned on the basis of height with limited accuracy. A more satisfactory approach corresponds to the method of Covell⁽²⁰⁾ developed originally for NaI spectra in which peaks are fitted by Gaussian functions to obtain resolved areas. If, however, a small peak resides on the shoulder of a larger one, the calculated net peak area of the smaller member is sensitive to the functional representation of the larger peak in the vicinity of the shoulder. At this laboratory a skewed Gaussian function combined with a linear background term has been used with some success in this type of situation but the increased running time in routine analysis makes it desirable to use alternative isolated peaks whenever possible. Fortunately, in Ge(Li) spectra, overlapping peaks are the exception rather than the rule so that alternative peaks can usually be found.

The correspondence between peak position and gamma energy, typically expressed in terms of an N^{th} order polynomial, enables the computer to locate peak positions if gamma energies are supplied or to identify radioisotopes possibly contributing counts under a given peak by various detection processes. From a knowledge of the experimental conditions it is generally obvious which isotope is primarily responsible for the presence of a given peak, but a thorough search of a gamma energy-isotope library can be helpful in identifying possible interferences which might otherwise be overlooked. In routine analyses of spectra with the same set of peaks recurring, it is of course unnecessary to use a peak identification option with each spectrum.

Net peak areas must be converted to isotope weights by means of appropriate factors which include a correction for the reduction in the number of counts due to the dead time of the multichannel analyzer. Most modern multichannel analyzers have "live time" clocks which correct for instrumental dead time by extending real time counting periods. Gavron⁽²¹⁾ has pointed out that in addition to the fact that these clocks are not always accurate, they can not make the proper dead-time compensation for short lived samples where no simple relation exists between the real time and the fractional dead time. An algorithm has been developed⁽²¹⁾ to handle this problem but it is desirable to keep counting rates sufficiently low that corrections having only approximate validity are unnecessary. In addition to conversion of net peak areas, the computer should make appropriate cooling and counting time decay corrections, flux normalizations, subtractions for interferences as indicated in Ch. 2 and calculate confidence levels in reported values.

It is readily seen that hand extraction of the useful information content of gamma spectra is a tedious and time consuming endeavor subject to a variety of human errors. When spectral data are complex or when a large number of spectra must be analyzed, as in the case of environmental sampling, it is especially desirable that data reduction be accomplished efficiently via computer processing with minimal human intervention. Because of the complexity of gamma spectra, computer programs have not yet been successful in taking over the entire job of data reduction, but the greatest share of the work can in fact be done automatically.

Numerous computer-based spectrum analysis methods are reported in the literature, especially for use with NaI spectra. The FORTRAN IV programs developed here for analysis of Ge(Li) spectra obtained from radionuclide mixtures present in suites of irradiated^c environmental samples, provide a broad data reduction capability. The operations outlined in Fig. 10 are written in terms of subroutines which may be incorporated into main programs meeting the specialized requirements of the investigator.

In the following section, details connected with storage of spectral data on magnetic tape and its subsequent retrieval by the computer are considered. Sufficient procedural information is presented to make this section hopefully a useful guide on how to get data from the analyzer into the University of Michigan IBM 360/67 computer. In section 3 the available subprograms and short main programs are described while in the final section the main program in current use with the analytical procedure outlined in Chapter 2 is described in detail.

2. Tape analysis:

A. Tape structure. Each spectrum is written on tape either as a set of N/256 records each of which is 258X6 BDC characters long or as one continuous record (N+2)X6 characters long where N=1024, 2048, 4096 channels. The writing mode is determined by a switch labeled (IRG/256) in the back of the ND 2200 series magnetic tape module. Existing programs expect the tape to be formatted in the latter way as one large record per spectrum. In this format the tagword is written twice at the beginning of the long record, while in the blocked format it is written twice at the beginning of each segment of 256 channels as shown in the ND 2200 series magnetic tape manual. For each channel, counts are represented on tape by a string of six BCD characters. Since tagword values have at most four digits, two of the six characters for the tagword contain no useful information.

B. Tape submission. Tapes should be submitted at the batch input window at the University of Michigan Computing Center, North University Building, Main Campus. Two labels are affixed physically to each reel submitted: (1) Computing Center ID (CCID) of the form Gxxx (e.g. G123) and (2) users tape ID (UTID) concocted for esoteric reasons by the user (e.g. SOURCE). In exchange for a tape the user receives a green card receipt having the appropriate Gxxx punched in the first columns. So as not to forget the association between the CCID and UTID it is desirable to write the UTID on the receipt card. Tapes so entered in the C.C. users library may be retrieved at the output window at a later time in exchange for the green card. Tapes may stay in the library so long as they are used at

least once a month. According to Computing Center personnel, tapes may not be retrievable if they are not used for several months or if the green card is lost! The UTID may be permanently associated with the tape while a new CCID will be assigned if the tape is withdrawn and subsequently resubmitted.

The green card itself is never used as part of a series of program input cards. The information about what tape to use is supplied on a \$RUN *MOUNT command card described below. For the protection of the user, the CCID and UTID are intercompared before the tape is mounted by the operator. If the association is incorrect, the tape will not be mounted.

C. Tape Inventory. Provided there are no parity or IRG "noise" errors on a tape, the string of BCD characters representing a spectrum can be translated into bytes and correctly grouped into integers giving counts per channel by the usual formatted FORTRAN read statement. Often the original tape has errors and very short records sometimes arising from user intervention in the write operation at the wrong moment. The simple read statement will not handle such troubles for, upon detection of errors, control is passed back to MTS.

A possible solution is to prepare an error-free tape using the MTS routine *TAPECOPY (Computing Center memo M 102) which can copy all records including those with parity errors. For faulty records the parity is re-calculated so as to be correct on the new tape. This program will not give information about the tagword and record length associated with each record.

The FORTRAN tape inventory program INVEN (p. 94) does have this capability but will reject any record which is not of correct parity. In fact the program as presently constituted will transcribe only records which are of correct parity, are "noise" free and of correct length (i.e. a multiple of 1024 channels). These more stringent requirements are adopted because it is not always clear how the record is in error if the parity is wrong or if the record is of improper size. An important advantage of having a tape inventory, is that it allows one to search

for a desired record on the basis of its position among many records rather than on the basis of its tagword. Even though it is desirable and customary to write record sequences with automatic tagword incrementing, it sometimes happens because of the human touch, that more than one record will have the same tagword, in which case record order is of clear importance.

The input card sequence for INVEN is shown on page 38.

Card 3: Beginning of MOUNT instructions.

Card 4: Mount tape with CCID G123 on a seven track unit (there are only two at the Computing Center), assign pseudo device name *R* to it and convert (N), 200 char. per inch (2), even parity (E) characters to bytes using a tape buffer storage size of 25,000 characters. The UTID is SOURCE. By default (i.e. omitting the statement ring = out) the tape ring is taken out which means no writing can be done on this tape (which is the original after all!) The Computing Center operator will put in or remove rings as per instruction on the \$RUN *MOUNT card. For additional information on *MOUNT consult Computing Center memo M67, M68 and CC News #145. (22)

Card 5: Same as 3 but one writes on this tape. Hence ring = IN. UTID = CLTP6

Card 6: End of MOUNT instructions.

Card 7: Load and run the compiled main program +MVL in the file INVEN. FORTRAN uses integers instead of pseudo device names; hence the correspondence, 2=*R*, 4=*W*, is made at load time. The program reads data from unit 2 and writes on tape mounted on unit 4.

Card 8: Name of the tape, perhaps UTID, written on new tape inventory sheet. (2A4) format.

Card 9: Any additional information to be printed out on the new tape inventory sheet (20A4 format).

Card 10: N, NSK in 2I4 format.

The inventory begins after NSK records on the source tape have been skipped

and continues transcribing through record with tagword N before quitting. In the automatic incrementing mode this number can usually be equal to the last tagword written on the tape. If automatic incrementing is not used, a dummy spectrum should be written at the very end with N=9999.

Program Notes: Details can be understood by inspection of the FORTRAN listing (p. 94). A unformatted read subroutine is used in which the modifier m is defined (hex: z400000000) so as to transmit error messages to the program rather than to MTS. A detected error will result in repeated read-back-space operations by the tape drive. This behavior is curtailed by invoking a one record SKIP operation (Computing Center memo M80) whenever more than four unacceptable files (too short usually), four end-of-file marks or two permanent errors are detected in succession. This apparently arbitrary approach has greatly decreased the inventory time without impairing the efficiency of record retrieval. The most frequently encountered error is parity, code = 16. In the case of a tape drive failure, code = 24, the inventory will terminate. Experience has shown that if this error occurs, one should merely resubmit the job without changing any cards. Usually the second time around this error, which may arise because of the marginal compatibility of the Kennedy and the 360 tape drives, does not occur. Fortunately the code 24 error rarely occurs.

Having produced a new tape, the program rewinds it and inspects it in essentially the same way for errors. Thus an error-free inventory of this tape indicates that all is well for subsequent data analysis. Cost of the inventory: 10¢/4096 channels.

3. Output: Part of a source inventory is shown on page 40. EOF=End of file, FLN=File number, LEN=number of characters in the record, CHNLS= corresponding number of channels, CMRD=cumulative number of read operations, CRN=index of acceptable records written on new tape. Of the 42 records on this tape only one (Tg=0039) was found to be unacceptable. Thus the inventory of the new tape also

Input cards for Running the
compiled Fortran Program INVEN.

1. \$SIGNON SN87 T=IM 'NAME'
2. PASSWORD
3. \$RUN *MOUNT
4. G123 ON 7TP *R* MODE=2EN SIZE=25000 'SOURCE'
5. G308 ON 7TP *W* MODE=2EN RING=IN SIZE=2500 'CLTP6'
6. \$ENDFILE
7. \$RUN INVEN 2=*R* 4=*W*
8. CLTP6
9. WRITTEN ON 9/26/69
10. 00420000
11. \$SIG

listed on p. 40 displays the tagword and record length for 41 records. All extra end-of-file marks between records have been deleted.

Shown on p. 41 is part of a source inventory for an unusually bad case. The program detected two parity errors in a row after Tg=0041, decided thus to skip one record, immediately found a record far too short followed by another record with incorrect parity. After skipping a region with too many end of file returns, it detected a series of extremely short records which in the case of "Tg"=0213 it attempted to read 3 times before invoking skip. Notice that despite patches of trouble, no useful information was lost. The records were written in the automatic incrementing mode and each tagword is in fact present on the transcribed tape (CRN=14). Occasionally a desired spectrum is lost, as in the previous example (Tg 0039). The ratio of lost to saved spectra is about 1/50. It is desirable to write very important spectra twice to insure their retrievability. The Kennedy tape recorder accepts 1200 foot reels which can store about 100 4096 channel spectra. Thus one reel will take about 50 duplicate spectra of this size.

3. Data Analysis Programs.

Large multipurpose programs are built out of collection of subroutines having specific tasks, thus permitting rapid assembly of programs to fit specialized needs. In this section the subroutines are described as well as several short main programs which make use of them.

A. Tape read and list (RDLST). A set of routines read the tape (either the new tape produced by the inventory program or an error free original tape) and print out the contents of each channel. The main program (p. 95) (RDLST) makes use of five subroutines RDTAPE, MVL, WRTMT, BASE, and WRTBS. As seen from the FORTRAN listing of RDTAPE (p.95) the tape read format is 3A4, 32(128I6). Reading in the first 12 characters as 4 byte words avoids complications connected with the special encoding of the tagwords. Namely:

Inventory Program Output :

CONTENTS OF TAPE BEING READ

CONTENTS OF NEWLY WRITTEN TAPE CLTR

WRITTEN ON 9/20/85.

ECF 1
ECF 2
ECF 3
ECF 4
ECF 5
ECF 6
ECF 7
ECF 8
ECF 9
ECF 10
ECF 11
ECF 12
ECF 13
ECF 14
ECF 15
ECF 16
ECF 17
ECF 18
ECF 19
ECF 20
ECF 21
ECF 22
ECF 23
ECF 24
ECF 25
ECF 26
ECF 27
ECF 28
ECF 29
ECF 30
ECF 31
ECF 32
ECF 33
ECF 34
ECF 35
ECF 36
ECF 37

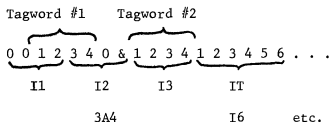
FLN 1 TOW C001 LEN 12303 CHMLS 2048 CMDR 3 CRN 1
FLA 2 TOW C002 LEN 12300 CHMLS 2040 CMDR 6 CRN 2
FLN 3 TOW C003 LEN 12300 CHMLS 2048 CMDR 9 CRN 3
FLN 4 TOW C004 LEN 12330 CHMLS 2048 CMDR 12 CRN 4
FLN 5 TOW C005 LEN 12300 CHMLS 2048 CMDR 15 CRN 5
FLN 6 TOW C006 LEN 12300 CHMLS 2048 CMDR 18 CRN 6
FLN 7 TOW C007 LEN 12300 CHMLS 2048 CMDR 21 CRN 7
FLN 8 TOW C008 LEN 12309 CHMLS 2048 CMDR 24 CRN 8
FLN 9 TOW C009 LEN 12390 CHMLS 2048 CMDR 27 CRN 9
FLN 10 TOW C010 LEN 12300 CHMLS 2048 CMDR 30 CRN 10
FLN 11 TOW C011 LEN 12300 CHMLS 2048 CMDR 33 CRN 11
FLN 12 TOW C012 LEN 12300 CHMLS 2048 CMDR 36 CRN 12
FLN 13 TOW C013 LEN 12330 CHMLS 2048 CMDR 39 CRN 13
FLN 14 TOW C014 LEN 12300 CHMLS 2048 CMDR 42 CRN 14
FLN 15 TOW C015 LEN 12300 CHMLS 2048 CMDR 45 CRN 15
FLN 16 TOW C016 LEN 12300 CHMLS 2048 CMDR 48 CRN 16
FLN 17 TOW C017 LEN 12300 CHMLS 2048 CMDR 51 CRN 17
FLN 18 TOW C018 LEN 12300 CHMLS 2048 CMDR 54 CRN 18

FILE AD. 1 TOW C001 LEN 12300 CHMLS 2048 CMDR 3 CRN 1
FILE AD. 2 TOW C002 LEN 12300 CHMLS 2048 CMDR 6 CRN 2
FILE AD. 3 TOW C003 LEN 12300 CHMLS 2048 CMDR 9 CRN 3
FILE AD. 4 TOW C004 LEN 12300 CHMLS 2048 CMDR 12 CRN 4
FILE AD. 5 TOW C005 LEN 12300 CHMLS 2048 CMDR 15 CRN 5
FILE AD. 6 TOW C006 LEN 12300 CHMLS 2048 CMDR 18 CRN 6
FILE AD. 7 TOW C007 LEN 12300 CHMLS 2048 CMDR 21 CRN 7
FILE AD. 8 TOW C008 LEN 12309 CHMLS 2048 CMDR 24 CRN 8
FILE AD. 9 TOW C009 LEN 12390 CHMLS 2048 CMDR 27 CRN 9
FILE AD. 10 TOW C010 LEN 12300 CHMLS 2048 CMDR 30 CRN 10
FILE AD. 11 TOW C011 LEN 12300 CHMLS 2048 CMDR 33 CRN 11
FILE AD. 12 TOW C012 LEN 12300 CHMLS 2048 CMDR 36 CRN 12
FILE AD. 13 TOW C013 LEN 12330 CHMLS 2048 CMDR 39 CRN 13
FILE AD. 14 TOW C014 LEN 12300 CHMLS 2048 CMDR 42 CRN 14
FILE AD. 15 TOW C015 LEN 12300 CHMLS 2048 CMDR 45 CRN 15
FILE AD. 16 TOW C016 LEN 12300 CHMLS 2048 CMDR 48 CRN 16
FILE AD. 17 TOW C017 LEN 12300 CHMLS 2048 CMDR 51 CRN 17
FILE AD. 18 TOW C018 LEN 12300 CHMLS 2048 CMDR 54 CRN 18
FILE AD. 19 TOW C019 LEN 12300 CHMLS 2048 CMDR 57 CRN 19
FILE AD. 20 TOW C020 LEN 12300 CHMLS 2048 CMDR 60 CRN 20
FILE AD. 21 TOW C021 LEN 12300 CHMLS 2048 CMDR 63 CRN 21
FILE AD. 22 TOW C022 LEN 12300 CHMLS 2048 CMDR 66 CRN 22
FILE AD. 23 TOW C023 LEN 12300 CHMLS 2048 CMDR 69 CRN 23
FILE AD. 24 TOW C024 LEN 12300 CHMLS 2048 CMDR 72 CRN 24
FILE AD. 25 TOW C025 LEN 12300 CHMLS 2048 CMDR 75 CRN 25
FILE AD. 26 TOW C026 LEN 12300 CHMLS 2048 CMDR 78 CRN 26
FILE AD. 27 TOW C027 LEN 12300 CHMLS 2048 CMDR 81 CRN 27
FILE AD. 28 TOW C028 LEN 12300 CHMLS 2048 CMDR 84 CRN 28
FILE AD. 29 TOW C029 LEN 12300 CHMLS 2048 CMDR 87 CRN 29
FILE AD. 30 TOW C030 LEN 12300 CHMLS 2048 CMDR 90 CRN 30
FILE AD. 31 TOW C031 LEN 12300 CHMLS 2048 CMDR 93 CRN 31
FILE AD. 32 TOW C032 LEN 12300 CHMLS 2048 CMDR 96 CRN 32
FILE AD. 33 TOW C033 LEN 12300 CHMLS 2048 CMDR 99 CRN 33
FILE AD. 34 TOW C034 LEN 12300 CHMLS 2048 CMDR 102 CRN 34
FILE AD. 35 TOW C035 LEN 12300 CHMLS 2048 CMDR 105 CRN 35
FILE AD. 36 TOW C036 LEN 12300 CHMLS 2048 CMDR 108 CRN 36
FILE AD. 37 TOW C037 LEN 12300 CHMLS 2048 CMDR 111 CRN 37
FILE AD. 38 TOW C038 LEN 12300 CHMLS 2048 CMDR 114 CRN 38
FILE AD. 39 TOW C039 LEN 12300 CHMLS 2048 CMDR 117 CRN 39
FILE AD. 40 TOW C040 LEN 12300 CHMLS 2048 CMDR 120 CRN 40
FILE AD. 41 TOW C041 LEN 12300 CHMLS 2048 CMDR 123 CRN 41
FILE AD. 42 TOW C042 LEN 12300 CHMLS 2048 CMDR 126 CRN 42
FILE AD. 43 TOW C043 LEN 12300 CHMLS 2048 CMDR 129 CRN 43
FILE AD. 44 TOW C044 LEN 12300 CHMLS 2048 CMDR 132 CRN 44
FILE AD. 45 TOW C045 LEN 12300 CHMLS 2048 CMDR 135 CRN 45
FILE AD. 46 TOW C046 LEN 12300 CHMLS 2048 CMDR 138 CRN 46
FILE AD. 47 TOW C047 LEN 12300 CHMLS 2048 CMDR 141 CRN 47
FILE AD. 48 TOW C048 LEN 12300 CHMLS 2048 CMDR 144 CRN 48
FILE AD. 49 TOW C049 LEN 12300 CHMLS 2048 CMDR 147 CRN 49
FILE AD. 50 TOW C050 LEN 12300 CHMLS 2048 CMDR 150 CRN 50

* SHORT RECORD:

A Source Tape Listing of an Unusually Bad Tape :

CONTENTS OF TAPE BEING READ										
EOF 1	FLN 1	TGW 0035	LEN 24588	CHNLS 4096	CMRD 1	CRN 1				
EOF 2	FLN 2	TGW 0036	LEN 24588	CHNLS 4096	CMRD 4	CRN 2				
EOF 3	FLN 3	TGW 0037	LEN 24588	CHNLS 4096	CMRD 7	CRN 3				
EOF 4										
EOF 5	FLN 4	TGW 0038	LEN 24588	CHNLS 4096	CMRD 10	CRN 4				
EOF 6										
EOF 7	FLN 5	TGW 0039	LEN 24588	CHNLS 4096	CMRD 13	CRN 5				
EOF 8										
EOF 9										
EOF 10										
EOF 11	FLN 6	TGW 0040	LEN 24588	CHNLS 4096	CMRD 17	CRN 6				
EOF 12										
EOF 13										
EOF 14	FLN 7	TGW 0041	LEN 24588	CHNLS 4096	CMRD 21	CRN 7				
ERR 1	CD 16									
ERR 2	CD 16	SKIP								
EOF 15										
EOF 16	FLN 8	TGW 0042	LEN 317	CHNLS 50	CMRD 25					
EOF 17	FLN 9	TGW 0042	LEN 24588	CHNLS 4096	CMRD 27	CRN 8				
EOF 18										
EOF 19	FLN 10	TGW 0043	LEN 24588	CHNLS 4096	CMRD 31	CRN 9				
ERR 3	CD 16									
ERR 4	CD 16	SKIP								
EOF 20										
EOF 21										
EOF 22										
EOF 23	SKIP									
EOF 24	FLN 11	TGW 0044	LEN 24588	CHNLS 4096	CMRD 38	CRN 10				
EOF 25										
EOF 26	FLN 12	TGW 0045	LEN 24588	CHNLS 4096	CMRD 41	CRN 11				
EOF 27	FLN 13	TGW 0046	LEN 24588	CHNLS 4096	CMRD 43	CRN 12				
EOF 28										
EOF 29										
	FLN 14	TGW 0047	LEN 24588	CHNLS 4096	CMRD 47	CRN 13				
	FLN 15	TGW 0002	LEN 36	CHNLS 4	CMRD 48					
	FLN 16	TGW 0213	LEN 27	CHNLS 2	CMRD 49					
	FLN 17	TGW 0213	LEN 27	CHNLS 2	CMRD 50					
	FLN 18	TGW 0213	LEN 27	CHNLS 2	CMRD 51					
QUEER FILE SKIP										
EOF 30	FLN 19	TGW 0048	LEN 81	CHNLS 11	CMRD 52					
	FLN 20	TGW 0048	LEN 24588	CHNLS 4096	CMRD 54	CRN 14				



(I3 = 1234) thus is the correct tagword in A4 format. The short subroutine MVL (p. 95) converts I3 to I format so that tagword input information from cards is conveniently compared. Subroutine RDTAPE then will read one record of length NR+1 channels. NR (e.g.: 1023, 2047 or 2095) must be supplied by the program while the tagword, I3 (in alphameric mode), the [^{live}_{actual}] time of data accumulation, IT, and the data array IR(J) (J=1,NR) are returned. IT is stored in the "zeroth" channel so that only NR channels contain pulse height data. Subroutine WRITMT (p. 95) will print out the values I3, IT and (IR(J), J=1, NR) obtained previously. BASE and WRTBS are described below.

The input card sequence required to print out portions of five of the forty-one spectra listed in the inventory of CLTR6 p. 44 are shown below. Six values entered on one card are needed for each spectrum listed; NTG, NREC, NSK, IS, IE and NBS in 614 format. NTG = the tagword sought, NREC = the total number of channels in the spectrum (i.e. 1024, 2048, or 4096), NSK = the number of records to be skipped starting with the present position of the tape, before searching for the target record (i.e. that with the desired tagword). The search operation consists in successive formatted reads and comparison of the NTG with the tagword read, I3. If the tagword sought is less than the tagword read, a mismatch is detected and the program will stop. This feature can easily be changed so that the tape will backspace an appropriate number of records in its searching but this would be desirable only if automatic incrementing has been completely successful. In this connection it is important to note that the operation of skipping records is much faster than the formatted read operation. Hence it is desirable to choose NSK so that the very next record is the target record. This can be

done on the basis of the tape inventory which gives the correspondence between the record index and tagword on the newly written tape. IS and IE are respectively the first and last channels of the spectrum to be printed out. The listing gives 500 channels per page of output. $NBS \leq 0$ for a simple listing. The case for $NBS > 0$ is discussed below.

The first card in the data set, Card 7, causes the last 1048 channels of spectrum with tagword 1 to be printed out. Card 8 causes the first 2000 channels of the spectrum with tagword 7 to be printed. In this case 5 spectra are skipped prior to searching for 7. As mentioned previously if NSK had been taken = 0 the same output would have been obtained but the running time would have been increased because spectra 2 through 6 would have been read in in order to find 7. A portion of the first page of output for a spectrum is shown below. The time is labeled "live-time" by the program in as much as this is the common operational mode of the analyzer but the actual meaning of this number is of course determined by the position of the live-time switch on the analyzer itself. The effects of cards 9 and 10 can be seen by inspection of the inventory on p. 40. A blank card will cause RDLST to stop.

B. Base spectrum generation and peak integration. The subroutine BASE (p.95) provides a measure of the pulse height spectrum baseline on which peaks rest, thus enabling the computer to make a determination of net peak areas. The technique is described in detail elsewhere (Ralston & Wilcox 19) but, briefly, the program generates from the original spectrum, S^0 , with n_i counts in the i th channel, a smoothed spectrum \bar{S}^0 in which the number of counts in the i th channel, \bar{n}_i , is taken as the average over an interval of width $2\ell+1$ centered about i .

Thus

$$\bar{n}_i = \frac{1}{2\ell+1} \sum_{j=i-\ell}^{j=i+\ell} n_j$$

Card inputs to RDLST

1. \$SIGNON SN87T=IM
2. PASSWORD
3. \$RUN *MOUNT
4. G308 ON 7TP *R* MODE=2EN RING=OUT SIZE=25,000 'CLTP6'
5. \$ENDFILE
6. \$RUN RDLST 4 = *R*
7. 000120480000100020480000
8. 000720480005000120000000
9. 003820480032050010000000
10. 004020480000000120000000
11. 004220480001070009000030
12. BLANK CARD
13. \$SIG

The First Page of Output from RDLST .

14WGR00= 00.16 LIVE TIME= 1000 SEC												
Channel												
Start	Data											End Channel
1	2	3	4	5	6	7	8	9	10	11	12	13
11	18	29	37	49	81	118	144	193	224	3	8	10
11	403	615	644	598	604	559	504	502	504	504	504	504
21	403	615	644	598	604	559	504	502	504	504	504	504
31	403	615	644	598	604	559	504	502	504	504	504	504
41	505	465	439	437	463	432	408	414	435	455	454	50
51	450	419	422	436	437	444	444	437	449	435	50	
61	450	419	422	436	437	444	444	437	449	435	50	
71	331	369	355	377	332	362	403	418	421	376	89	
81	324	331	332	320	364	326	295	342	329	315	90	
91	314	346	377	318	323	214	317	320	319	342	110	
101	328	322	321	341	340	316	335	309	311	323	120	
111	268	306	307	355	319	309	309	315	334	299	140	
121	268	306	307	355	319	309	309	315	334	299	140	
131	301	299	290	298	283	283	286	308	259	290	150	
141	301	299	290	298	283	283	286	308	259	290	150	
151	324	313	303	283	272	260	285	312	296	266	160	
161	254	254	276	270	245	238	237	249	267	255	200	
171	254	254	276	270	245	238	237	249	267	255	200	
181	251	270	264	264	251	287	240	268	274	247	190	
191	254	254	276	270	245	238	237	249	267	255	200	
201	254	254	276	270	245	238	237	249	267	255	200	
211	257	247	238	231	253	239	259	224	218	233	220	
221	229	248	240	255	242	269	221	211	237	232	230	
231	213	212	205	202	187	204	219	201	199	200	230	
241	199	187	198	166	175	184	175	165	175	187	194	
251	202	215	190	210	191	171	174	174	197	194	260	
261	185	161	181	181	170	155	171	156	177	175	290	
271	185	161	181	181	170	155	171	156	177	175	290	
281	185	161	181	181	170	155	171	156	177	175	290	
291	185	161	181	181	170	155	171	156	177	175	290	
301	156	171	184	156	169	171	169	181	197	137	320	
311	156	171	184	156	169	171	169	181	197	137	320	
321	170	161	166	168	163	170	157	162	154	144	330	
331	161	159	157	164	170	156	154	151	150	150	360	
341	152	155	136	159	154	138	140	142	142	142	360	
351	152	155	136	159	154	138	140	142	142	142	360	
361	154	157	162	134	130	152	153	137	156	142	370	
371	154	157	162	134	130	152	153	137	156	142	370	
381	147	156	156	143	126	133	135	149	142	161	390	
391	123	127	142	139	137	140	132	123	118	135	400	
401	123	127	142	139	137	140	132	123	118	135	400	
411	123	127	142	139	137	140	132	123	118	135	400	
421	105	127	126	128	131	129	151	139	104	129	430	
431	133	114	122	126	123	128	125	101	160	162	460	
441	133	114	122	126	123	128	125	101	160	162	460	
451	123	117	125	125	135	119	133	109	122	125	460	
461	122	122	123	131	139	98	116	117	124	108	470	
471	110	119	129	130	114	128	113	122	103	115	490	
481	134	110	120	161	115	128	100	138	134	121	500	
491	128	116	110	160	125	123	118	148	170	510		
501	128	116	110	160	125	123	118	148	170	510		
511	109	109	119	111	122	123	125	116	118	117	530	
521	109	109	119	111	122	123	125	116	118	117	530	

S^0 and $\overline{S^0}$ are compared. Whenever n_i exceeds $\overline{n_i}$ by a standard deviation (i.e. by $\alpha \sqrt{\overline{n_i}}$), n_i is replaced by $\overline{n_i}$ thus generating a new spectrum S^1 .

The above steps of smoothing and replacement are repeated in turn using S^1 in place of S^0 and so on for n cycles with ever-diminishing values of α . ($\alpha_{i+1} < \alpha_i$). The resultant spectrum S^n is taken as the baseline. In subroutine BASE the initial value of α is taken as 9.2, decreasing in ten (i.e. $n=10$) equally spaced steps to 0.2. ℓ is taken to be 30 channels. The choice for ℓ results from a compromise between the use of a small ℓ (~ 10 channels) for which the Compton edges are well-followed and a large value which gives a better representation of the baseline under unusually broad peaks. An example of the effects dependent on the choice of ℓ is shown in Fig. 11. The spectrum was taken at a very high counting rate in order to exaggerate peak widths. With $\ell=30$ channels, the calculated baseline is largely insensitive to peak width variations encountered in actual measurements.

In the plot of a typical Ge(Li) gamma Spectrum (Fig. 12). The smooth curve follows the upper limit of the calculated baseline. Portions of the spectrum which do not exceed $0.2 \overline{n_i}$ are not replaced in forming the baseline. Thus only under the peaks is the baseline smooth. Elsewhere it often has the value of the original data. The first and last ($2\ell+1$) channels (61 in this case) are lost in the analysis. It will be seen that the baseline follows well the curve that would be drawn by eye except in the vicinity of Compton edges. While this program provides a means for doing routine analysis of peaks known to be away from Compton edges, it is the responsibility of the user to insure that this is in fact the case for his particular experimental conditions. Also note that BASE tends to underestimate the "baseline" associated with small peaks on the shoulder of a large one as in the case of peak labeled 10 situated on the shoulder of peak 11.

Arguments of subroutine BASE (p. 95):

L = sum half width = 30

IST = starting channel (usually = 1)

IND = final channel (usually = NR)

IR = data array

BS = returned base spectrum array

The subroutine PKBS listed on p.95, "integrates" the peak simply by summing counts over $2*NFW+1$ channels centered about channel ICH. Base spectrum counts obtained previously are summed over the same interval. The subroutine returns SBS= the net counts =

$$\sum_{J=ICH-NFW}^{J=ICH+NFW} \{IR(J) - BS(J)\} \equiv N - B$$

the standard deviation, $SD = \sqrt{N + B}$, and the ratio = SD/SBS . In the expression for SD it is assumed that B has the Poisson- statistical uncertainty one would associate with real counts. For real peaks of about 3KeV FWHM, when the energy calibration is 1KeV/channel, the ratio SD/SBS as a function of NFW has a minimum around $NFW=2$. In order to compensate for possible line shape variations, NFW is typically taken = 3 which slightly increases SD in general (a few percent increase). PKBS requires the "peak" channel, ICH, as input. Thus any drift compensation must be done before calling PKBS.

A subroutine WRTBS (p.95) operates like WRTMT but lists the values of the base spectrum for each channel as well as the original data. In the read-list program RDLST, if $NBS > 0$, a base spectrum will be calculated from channel IS, to IE with $L=NBS$ (usually = 30) and WRTBS rather than WRTMT will be called. A sample portion of WRTBS output is shown on (p.49) for the last spectrum on CLTP6. This spectrum is produced from card 11 in the input (p. 44) to RDLST. This print out was originally obtained in order to examine the behavior of the computed baseline in the vicinity of the 846.9 KeV Mn^{56} peak. It will be seen that the actual counts

in a given channel (e.g. the "peak" channel, 842) is the upper number of the pair (2144) while the lower one is the baseline count (261). Arguments of subroutine WRITS:

IS = starting channel (usually 1)
IE = final channel (usually NR)
BS = previously calculated base spectrum

C. Plotting. A makeshift program has been written to product plots like that shown in Fig. 4. Limited to six consecutive plots, the program requires 5 values for each spectrum: NREC, ITG, NCUT, NSK, NCR.

NREC = no. of channels
ITG = tagword desired
NSK = skip NSK records before looking for ITG
NCR = 0 do not calculate base
 >0 calculate base spectrum and plot
NCUT = no. of channels to be plotted
NREC = 0 stop

The channel axis must be ≤ 10 inches. For 4096 channels this gives $FT = .0025$. While the log counts axis can be up to 24 inches. But for a fit on $8\frac{1}{2}'' \times 11''$ paper, assuming 10^4 cts maximum counts in any one channel, $FT = 2$ gives a maximum height or $8'' = 2 \log 10^4$.

The plot shown in Fig. 4 was produced from the series of input cards shown on p. 50. For a description of the MTS control cards, obtain the MTS plotting manual⁽²³⁾.

D. Peak search routine.

A peak finding routine, PEAK, (p.97), is available which has been tested and found to work well except in the case of doublets. This subroutine which in turn calls a series of subroutines FKZ, CIJWZ, SFWZ, locates peaks using a second difference operator function described in detail by Mariscotti.⁽¹⁶⁾ A FORTRAN listing of a test main program, PKTST, and the above subroutines is on p. 97. The twenty-five

Partial Output from RDLST with BASE Spectrum Option (NBS=30)

761	279	308	308	291	310	281	268	250	289	265	770
	279	281	281	278	281	281	268	250	278	265	
771	295	307	247	276	267	288	261	252	302	292	780
	278	278	247	276	267	279	261	252	278	279	
781	248	279	279	289	255	301	282	312	279	307	790
	248	279	279	289	255	280	282	281	279	281	
791	328	306	274	285	280	277	314	306	267	280	800
	282	282	274	285	280	277	282	283	267	280	
801	277	274	308	294	293	272	270	256	265	297	810
	277	274	283	284	284	272	270	256	265	284	
811	291	297	285	300	330	401	422	371	302	280	820
	283	284	285	293	293	283	283	282	281	280	
821	305	310	311	300	306	289	308	294	285	326	830
	279	279	277	277	276	275	274	273	272	271	
831	337	337	313	303	296	307	362	435	492	870	840
	270	269	269	268	267	266	265	265	264	263	
841	1814	2144	1543	1344	1261	777	390	271	224	222	850
	262	261	260	259	257	256	255	253	224	222	
851	224	236	222	237	242	207	247	220	225	219	860
	224	236	222	237	242	207	242	220	225	219	
861	240	221	229	247	231	249	230	226	214	219	870
	240	221	229	234	231	232	230	226	214	219	
871	239	263	210	224	224	200	230	205	243	206	880
	227	228	210	224	224	200	221	205	220	206	

Input Card Listing for Plot Program.

```
$SIGNON SN87 T=IM
PASSWORD
$CREATE PLOT SIZE=1000
$RUN *MOUNT
G308 ON 7TP *W* MODE=2EN RING=OUT SIZE=25000 'CLTP6'
$ENDFILE
$RUN *FORTRAN
.
.
.
FORTRAN LISTING P. 96
.
.
.
$ENDFILE
$RUN-LOAD#++*SOURCE*+*PLOTSYS 4=*W* 9=PLOT
.
.
.
COMPILED SUBROUTINES (RDTAPE, MVL BASE)
.
.
.
$ENDFILE
4096000818000006000
BLANK CARD
$RUN *CCQUEUE PAR=PLOT
$RUN *PERMIT PAR=PLOT
$SIG
```


labeled lines shown in the plot (Fig. 12) were identified as peaks by the program. It will be seen that even a small peak on the shoulder of a large one is currently detected as in the case of pair 3-4 or 10 and 11. Also Compton edges are recognized and excluded on the basis of behavior of the second difference function. The sensitivity of peak detection is adjustable. In the present case, peaks labeled (A) and (B) were not identified but would have been if the sensitivity had been increased.

The peak finding routines have not as yet been incorporated into composite programs for routine use because in quantitative analysis of environmental samples the same set of peaks generally recur in each spectrum. Further information on this program is available on request.

E. Component Identification. The subroutine IDCOM (p.98) makes use of a list of 243 gamma-emitting nuclides and their energies⁽¹²⁾ to determine which nuclides may contribute by various detection processes to a gamma spectrum peak observed in a channel corresponding to the energy E. If the fwhm of the peak is $2\delta E$, a gamma ray of energy, E_γ , will contribute provided

$$E_\gamma = E \pm \delta E$$

for complete energy absorption in the detector ("main" peaks) or

$$E_\gamma - 511\text{KeV} = E \pm \delta E \text{ when } E > 1022 \text{ KeV}$$

for single-escape peaks or

$$E_\gamma - 1022 \text{ KeV} = E \pm \delta E \quad E > 1022 \text{ KeV}$$

for double-escape peaks, or provided two gammas from the same nuclide sum to the correct energy

$$E_{\gamma 1} + E_{\gamma 2} = E \pm \delta E.$$

The nuclide data is contained in a private file GAMLIB but is available on cards. It includes a complete list of slow-neutron produced (n, γ) nuclides as well as some fast-neutron produced species and background nuclides such as Cs¹³⁷ and K⁴¹. Actually the file contains two lists: (1) A main list with nuclides

index, name, half-life (min.) and up to five energies of the most intense gammas emitted (primary gammas) and (2) an auxiliary list containing up to 100 secondary gamma energies. The division into primary and secondary groups is somewhat qualitative following the classification of Adams and Dams.⁽¹²⁾

IDCOM requires a set of measured gamma energies, (ES, in KeV) the total number in the set, IPK, and the time (in minutes, TIME) elapsed between the end of irradiation and the start of counting. The subroutine expects to treat an entire spectrum. Thus if one finds no "main" peaks associated with the primary gammas of a certain nuclide, one certainly should not expect to find any S.E., D.E., sum or secondary peaks from that nuclide in the spectrum. This might not be true if certain peak energies were missing from the set. IDCOM first makes a list of those nuclides for which there is in each case at least one main peak found and for which the elapsed time is less than ten half-lives. Other nuclides are not considered further. For each nuclide on this list the subroutine calculates S.E., D.E. and sum energies from the primary gamma energies, obtains secondary peak energies from the auxiliary GAMLIB list and looks for equalities between these values and the energies $ES \pm \delta E$. Each nuclide name and coded detection process attributed to a given peak is indexed and stored for eventual transmission out of the subroutine. Finally for all j with $ES(j) < 100$ KeV, the subroutine looks for an $ES(j^1)$ satisfying the relation, $\frac{ES(j^1)}{ES(j)} = 1.132$, which holds for the $K\alpha$ and K_β X-ray pair. If such a pair is found, the Z of the associated atomic nucleus is given by

$$Z = f(E) = 28.553 + 0.9485 E - 0.003085E^2$$

The value of the energy spread, $\delta E \approx \frac{FWHM}{2}$, is obtained from a function subroutine EF(E). IDCOM Arguments: (values returned by IDCOM; not intermediate values.):

N(j) = number of nuclide-process attributes associated with the jth peak of energy

ES(j), NI(K,J) = process code associated with the Kth attribute and Jth peak. Thus:

NI = 1 main peak

2 double escape peak

3 single escape peak

4 sum peak

5 secondary peak

6 X-ray pair member

IDX(K,J) = Nuclide index $1 \leq \text{IDX} \leq 243$ associated with the K^{th} attribute and J^{th} peak. If NI = 6, IDX is not the isotope index, but rather the Z of the X-ray transition nucleus. $\text{IDX} = 0.5 + f(E)$.

NS(L, Loc) = Nuclide symbol for the L^{th} nuclide on the nuclide list. Two locations required for the symbol. LOC=1,2.

NPP = total number of nuclides in the list.

IP(L) = Nuclide index associated with the L^{th} nuclide. This value is needed for finding the correct secondary gamma energy list.

INDX(L) = Actual number of primary gammas (excluding the 511 KeV gamma) associated with the L^{th} nuclide.

NPKL (L) = Number of primary gammas (excluding 511 KeV) associated with the L^{th} nuclide for which a main peak was present. $\text{NPKL}(L) \leq \text{INDX}(L)$.

ER(L,k) = The primary gamma energies associated with the L^{th} nuclide. $K=1, 5$.

If $N(j) = 0$ the j^{th} peak is said to be "not attributed".

Subroutine WRTID (FORTRAN p.98). This subroutine will output information from IDCOM. It must be called for each peak, of energy ES(j). From a data array IZ, WRTID picks the correct atomic element symbol to go with the calculated Z when NI=6.

Although in typical use, IDCOM and WRTID would be used in connection with the peak finding routines PEAK etc. (p. 97) using values of ES obtained from it, a small main program IDTST (p. 98) exists which takes data from cards and provides a summary of isotopes possibly present.

PEAK NO. 1	ENERGY 74.1 KEV	PEAK NO. 17	ENERGY 2146.6 KEV		
12 T0141 PAIR PEAK		3 CL30 PAIR PEAK			
14 T1177 PAIR PEAK					
21 T0230 B-RAY PEAK Pu		PEAK NO. 18	ENERGY 2242.0 KEV		
		1 NA24 SINGLE ESCAPE PEAK			
PEAK NO. 2	ENERGY 85.2 KEV	PEAK NO. 19	ENERGY 2522.0 KEV		
11 EL155 PAIR PEAK		5 MU56 SPALL PEAK			
16 T0170 PAIR PEAK					
20 T0233 B-RAY PEAK Pb					
PEAK NO. 3	ENERGY 346.0 KEV	PEAK NO. 20	ENERGY 2651.0 KEV		
9 EU152 MAIN PEAK		5 MU56 SUP PEAK			
10 EU152 PAIR PEAK		5 MU56 SPALL PEAK			
12 CO154 PAIR PEAK					
14 EU152 PAIR PEAK					
16 T1177 PAIR PEAK		PEAK NO. 21	ENERGY 2755.0 KEV		
20 T0233 B-RAY PEAK Pb		1 NA24 PAIR PEAK			
24 NA24 SECURE ESCAPE PEAK					
PEAK NO. 4	ENERGY 511.0 KEV	PEAK NO. 22	ENERGY 2962.0 KEV		
8 SE23 PAIR PEAK		5 MU56 SPALL PEAK			
17 CO154 PAIR PEAK					
19 T1202 PAIR PEAK					
22 EU152 PAIR PEAK		PEAK NO. 23	ENERGY 3060.0 KEV		
16 T1177 PAIR PEAK		3 CL30 SLW PEAK			
20 T0233 B-RAY PEAK Pb					
PEAK NO. 5	ENERGY 847.0 KEV				
5 MU56 PAIR PEAK					
24 T1124 PAIR PEAK					
PEAK NO. 6	ENERGY 1144.0 KEV				
23 SE23 PAIR PEAK					
3 CL30 SECURE ESCAPE PEAK					
PEAK NO. 7	ENERGY 1173.1 KEV				
1 4 NA24	2 2 1308.9	2793.9	0.0	0.0	0.0
2 6 ALP20	1 1 1776.0	0.0	0.0	0.0	0.0
3 10 CL30	2 2 1642.0	4146.0	0.0	0.0	0.0
4 12 K42	3 1 1394.7	0.0	312.9	104.0	0.0
5 23 MU56	3 3 846.9	1810.7	2112.8	0.0	0.0
6 27 CO40	2 2 1332.4	1332.4	0.0	0.0	0.0
7 28 CO60	2 1 995.0	0.0	1132.4	0.0	0.0
8 30 SE23	5 1 106.6	226.9	515.0	717.4	750.7
9 150 EU152	5 1 346.2	1407.9	121.8	776.6	984.1
10 151 EU152M	5 1 461.6	121.8	903.3	344.2	1115.0
11 153 EU155	3 1 86.6	105.7	40.5	44.0	52.0
12 155 EU155	4 1 305.5	0.0	307.5	223.4	51.4
13 156 T1601	4 1 34.9	40.1	57.7	84.1	0.0
14 166 T1710	1 1 84.4	0.0	0.0	0.0	0.0
15 176 HF101	5 1 682.2	133.1	345.7	426.0	619.0
16 182 W187	5 1 445.7	434.3	72.1	134.3	93.1
17 188 CO190P	5 1 616.4	104.7	361.2	502.0	510.0
18 206 T2197P	3 1 356.3	0.0	0.0	0.0	0.0
19 207 T1202	3 1 440.2	0.0	521.3	309.4	0.0
20 209 T1633	5 1 44.6	459.2	185.3	182.2	070.0
21 211 W1919	1 1 74.7	0.0	0.0	0.0	0.0
22 214 W1618	3 1 911.9	612.4	1040.5	0.0	0.0
23 228 SE23	5 1 1302.3	241.0	435.9	553.0	1340.0
24 233 T1134	1 1 866.7	863.0	421.6	1072.4	109.1
25 241 B1411	5 1 189.6	276.9	304.9	344.7	457.9
PEAK NO. 10	ENERGY 1526.7 KEV				
4 SE23 PAIR PEAK		0 ON TERMINATED			
PEAK NO. 11	ENERGY 1642.0 KEV				
5 CL30 MAIN PEAK					
PEAK NO. 12	ENERGY 1655.0 KEV				
3 CL30 SINGLE ESCAPE PEAK					
PEAK NO. 13	ENERGY 1731.0 KEV				
1 NA24 SECURE ESCAPE PEAK					
24 T1134 SUP PEAK					
PEAK NO. 14	ENERGY 1778.6 KEV				
2 ALP20 PAIR PEAK					
PEAK NO. 15	ENERGY 1810.5 KEV				
5 MU56 PAIR PEAK					
PEAK NO. 16	ENERGY 2132.0 KEV				
5 MU56 PAIR PEAK					

Output from IDCOM Test Program IDTST.

Card input is evident from the FORTRAN listing (p. 98). The first card contains the "cooling" time in minutes. Subsequent cards contain one gamma energy each (in KeV). Up to 100 energies may be entered. The last card in this series should be blank. Sample output is shown on page 54. Each peak is first listed separately with its attributes. Thus peak 3 has seven possibly contributing nuclides. The larger number of attributes at low energies is in part due to the assumption of a constant peak FWHM of $2\delta E = 4\text{KeV}$. A form used in later runs was

$$\delta E = 0.7 + .001E \text{ (KeV)}$$

IDTST provides a summary table in the form of a list of elements found, with the number of main peaks in the library compared with those found in the actual spectrum. The print out also includes the main gamma energies in the library. This table is helpful in determining which elements are really present. Generally speaking if not all main peaks are present for a given isotope its presence may be doubted.

F. Non-linear least squares routine.

Let the set (Y_j, X_j) $j = 1, N$ be N observations on Y and X which are presumed to be related by $Y_j = f(X_j, \beta_1 \dots \beta_m)$ where the β_i 's ($i = 1, M$) are M adjustable parameters. The subroutine NLLS determines a parameter set β_i , starting from initial guesses for β (say β_i^0), which minimizes the residual

$$R \equiv \sum_{j=1}^N W_j (y_j - f(x_j, \beta_1 \dots \beta_m))^2$$

with respect to each β_i and thus produces a "best" fit according to this least squares criterion. That is, a set β_i is found such that $\frac{\partial R}{\partial \beta_i} = 0$ for all i . W_j is the weight attached to the j^{th} observation. If there is more than one parameter set which minimizes R , the solution obtained by the program will depend upon the starting point, i.e. the guesses for β_i . Moreover the functions must be bounded

in β_1^n in order to insure convergence. Despite such limitations the routine is of great usefulness and power.

Its application to the analysis of Ge(Li) spectra is two-fold.

1) Energy Calibration

2) Resolution of Doublets

The algorithm by which the minimization is accomplished is described briefly in Reference 24. Since a solution cannot be obtained in closed form generally, the equation is "linearized" by calculating small increments, δ_i , to initial values of β_i^0 , which will tend to minimize R, that is new values of β_i ,

$$\beta_i^1 = \beta_i^0 + \delta_i^0$$

are calculated for which

$$f(X_j, \beta_1^1, \dots) \approx f(X_j, \beta_1^0, \dots) + \sum_{i=1}^M \left. \frac{\partial f}{\partial \beta_i} \right|_{\beta_i = \beta_i^0} \delta_i^0$$

such that

$$\frac{\partial}{\partial \delta_i^0} \sum_{j=1}^N w_j (y_j - f(X_j, \beta_1^0, \dots))^2 - \sum_{i=1}^M \frac{\partial f}{\partial \beta_i} \delta_i^0 = 0$$

A solution of these M linear equations for each δ_i^0 allows one to correct β_i^0 in the direction of minimum R. The new values of β_i , β_i^1 , constitute new guesses for β_i which themselves may be corrected by calculation of a new δ_i^1 . This iterative procedure may be repeated until a desired accuracy in calculating each β_i is achieved.

That is, until

$$\left| \frac{\beta_i^n - \beta_i^{n-1}}{\beta_i} \right| \leq \epsilon$$

for all i where ϵ is small.

Equation can be seen to be a matrix equation. Defining $F_j = (Y_j - f(X_j, \beta_1, \dots))$ taking the partial with respect to δ_K , gives,

$$\sum_{j=1}^N w_j (F_j - \sum_{i=1}^M \frac{\partial f}{\partial \beta_i} \delta_i) \frac{\partial f}{\partial \beta_K} = 0$$

which gives

$$Y_K \equiv \sum_{j=1}^N w_j F_j \frac{\partial f}{\partial \beta_K} = \sum_{i=1}^M \sum_{j=1}^N w_j \frac{\partial f}{\partial \beta_i} \frac{\partial f}{\partial \beta_K} \delta_i \equiv A_{iK} \delta_i$$

where A_{iK} is a symmetric matrix with elements given by

$$\sum_{j=1}^N w_j \frac{\partial f}{\partial \beta_i} \frac{\partial f}{\partial \beta_K}$$

symbolically the solution for δ_i is then

$$\delta_i = A_{iK}^{-1} Y_K$$

where A_{iK}^{-1} is the inverse matrix associated with A_{iK} . The standard error associated with each parameter β_i is given by

$$\sigma_{\beta_i} = \sqrt{\frac{1}{N-M} \left(\sum_{j=1}^N w_j F_j^2 \right) |A_{iK}^{-1}|}$$

The arguments of the FORTRAN listing for NLLS are adequately described by the comment cards (p. 99). Matrix inversion is accomplished by the subroutine INV called from the UM function library. The functional relationship $f(X_j, \beta_1, \dots)$ is supplied by the user as the subroutine ARG. The fortran listing of ARG on p. 99 is for a power series,

$$f(X_j, \beta_1, \dots, \beta_m) = \beta_1 + \beta_2 X_j + \beta_3 X_j^2 \dots$$

Where U is the value of X_j for a specified J. ARG must be called for each data point. The dummy, P(1), contains the number of parameters = the polynomial order +1. V is the value of $f(X_j, \beta)$ returned. The current values of β are stored as G(I), I=1, M, and the derivatives, $\frac{\delta f}{\delta \beta_j}$, are returned by the subroutine as D(I) I=1, M. Thus a linear least squares fit would be obtained by using NLLS-ARG with P(1) = 2.

Since the correspondence between the energy of a detected gamma ray and the position of its peak in the analyzer spectrum is very nearly linear for the Nuclear Data 2200 unit, the addition of a small quadratic "correction" term suffices for most analytic work undertaken on this instrument. Thus a value of P(1) = 3 is customarily used but the capability exists of making higher-order fits for gamma energy determinations. Experience has shown that with reasonable initial guesses (within 20% of correct values) the quadratic fit using up to perhaps 10 lines, converges in 2 or 3 iterations with $\epsilon = 0.1\%$.

For purposes of energy calibration less powerful techniques of course, may suffice but, in addition, NLLS provides a means of analyzing doublets. Given the functional form, Gaussian or skewed Gaussian, of each of two peaks which are superposed NLLS can estimate their contributions under a broader range of conditions than may be done well by eye. Some experience has been gained in application of this program but its need in analysing typically encountered samples is as yet not great.

4. Composite Program

The program SPAN (FORTRAN p.100) is designed to obtain weights of elements present in samples deposited on various substrates and irradiated according to the analytical scheme discussed in Chapter 2. For each of the four combinations of irradiation and counting modes, the program is supplied with a list of gamma energies for which it estimates the position of corresponding peaks in the analyzer spectrum at hand and calculates net peak areas. The general features of the program sequence can be seen from the abbreviated flow diagram (Fig. 13.) A sample input card set is shown on p. 61. Data on these cards were developed and actually used in analysis of air particulate samples. Up to 50 gamma energies may be contained in each of the four blocks of isotope data accepted by the program. Two cards are required for each gamma energy entered. In the program the four run modes are indexed as follows:

ITY	Irradiation	Counting Live Time
1	5 minutes	400 sec
2	"	1000
3	5 hours	2000
4	"	4000

For the short irradiations (ITY=1,2) the isotope cards have the following format:

CARD	1	NS	E	SM	FK1	DF1	FK2	DF2
		2A4	2X	F10.5	F10.5	F10.5	F10.5	F10.5
	2	POL	DP	SOR	DS	POR	DR	
		F10.5	F10.5	F10.5	F10.5	F10.5	F10.5	

NS = the isotope symbol (e.g. CA49) (ITY=1)

E = gamma energy (KEV) (e.g. 3083.0 KEV)

SM = weight of element (i.e. isotopic mixture not the individual radionuclide) in the standard. (Arbitrary units e.g. 99.8 μ g)

FK1 = the factor converting net peak areas (counts) to element weight in units of SM based on some standard flux measure. (Say unit: 0.211 μ g/count)

DF1 = the standard error in FK1. (.0275 $\mu\text{g}/\text{count}$)

FK2, DF2 are similarly defined for a second counting arrangement differing from that used above. (e.g. FK2 determined for a larger distance between the sample and the detector).

It will be seen that as an analytical technique is developed FK1 and DF1 are not initially known. If the program is used with those values omitted, the output will consist of unconverted net peak areas. These areas can be used to obtain the FK's which can in turn be entered in the appropriate spaces on the card. The computer could have been programmed to make this calculation itself except that (1) once determined, FK's should not have to be re-computed, and (2) the estimate of DF should involve not only the Poisson-statistical uncertainty of individual estimates but also the uncertainties in reproducing the value of FK experimentally. Thus FK and DF should result from a set of measurements for which are most conveniently analyzed once by "hand" using the computer to provide net peak areas.

The second of the two isotope cards contains substrate data.

POL = the amount of element in units of SM present in the substrate on which the unknown is deposited per standard substrate area or volume. (i.e. $\mu\text{g Ca}$ on a full-circle polyethylene collector sheet.)

DP = the uncertainty in the value POL.

The pairs SOR, DS and POR, DR are similarly defined, so that the program as presently written allows substrate or blank subtraction to be made for three different materials. If no data is entered on this card, the program will operate properly assuming zero for blank subtraction. It will be seen that given FK1 or FK2, the activity of the blank material can be found initially by running the program for blanks as samples with zeros for the blank data. The weighted averages and standard deviations resulting from measurement of a number of duplicate blanks and analysis by computer, enables these zeros to be replaced.

A List of Input Cards to SPAN.

The order of isotopes within a block is generally unimportant but in its present version the program corrects the total counts from $Mg^{26}(n, \gamma)Mg^{27}$ activity ($E=1014$ KeV) for the interference arising from the $Al^{27}(n,p)Mg^{27}$ reaction. In order to make the correction properly in the present program, the Al isotope cards must appear third in block with ITY=1 and the Mg isotope cards must appear sixth on the block with ITY=2. Moreover the analysis of the ITY=1 spectrum must precede analysis of the ITY=2 spectrum.

For the long irradiations (ITY=3,4) the isotope cards contain the following data:

Single Isotope set	NS		E		SM		ASH		DA		TAU		FMY	
	1.	2A4	2X	F10.5	F10.5	F10.5	F10.5	F10.5	F10.5	F10.5	F10.5	F10.5	F10.5	F10.5
	2. Same as card 2 for ITY=1,2													

NS, E, SM have the same meaning as before. (Say Zn 69M (ITY=3), 438.7 KeV, 5.055 μ g).

ASH = the amount of element present in a standard-sized portion substrate containing the standard. (units of SM 0.1 μ g)

DA = the uncertainty in ASH (0.025 μ)

TAU = the isotope halflife in hours. (13.8 hr)

There are two choices for standards with ITY=3,4. Either SM or SM*FMY. The choice is determined by a parameter in the control cards described below.

The ordering of isotope cards is again unimportant except:

1) For ITY=3 the first card must be NA 1368.4 KeV and the second CU 511.0. The program corrects the observed 511 KeV counts arising mainly for the $Cu^{66}\beta^+$ annihilation for a small contribution arising from the energetic Na^{24} 2753.6 KeV gamma ray. Experimentally:

$$\text{Net CTS Due to } Cu^{66} = \text{total CTS} - 0.093 * \text{COUNTS in } 1368 \text{ KeV } Na^{24} \text{ peak}$$

2) For ITY=4 the first card must be SE75 264.6 KeV while the third must be Hg203 279.1. Since the 279.1 line is the only one available for Hg 203, it must be

used if amounts of this element is to be measured but the, 279.6 KeV Se75 line is virtually coincident with it. This interference can be removed by subtracting the counts inferred to be due to Se75 based on the ratio of counts in the 264.6 KeV and 279.6 KeV peaks for pure Se.

Experimentally $\text{Se}(279.6)/\text{Se}(264.6) = 0.385$ so that counts due to Hg = gross counts - $0.385 \times \text{CTS}$ from $\text{Se } 264.6$.

These interference corrections which presently necessitate ordering of the data and spectrum processing in the case of Mg-Al involve minor modifications of the program and can be removed if calculation of weights of these elements (Mg, Hg and Cu) are not of interest.

The end of each of the four isotope sets is defined by two blank cards.

The isotope data cards are followed by four corresponding sets of gamma energies (ITY=1-4) for use with an internal energy calibration routine. The list consists of energies corresponding to recurrent, prominent, isolated peaks. Energies entered may not necessarily be those in the isotope data set. While gross changes in the calibration must be treated by supplying information externally from cards it has proved useful to compensate for small alterations in calibration (shifts of up to several kilovolts) by means of information in the spectrum itself. Expected peak positions calculated using the above lists locate an interval over which a search is made for the absolute maximum. If the maximum occurs at the ends of the interval, it is considered out of range for inclusion in the final list of peaks for calibration. If it is within the range its statistical significance is checked. If it qualifies statistically, the centroid is located using a parabolic interpolation routine XM. This routine has the effect of adding the correct decimal point to the channel with maximum counts. The subset of energies and identified centroids satisfying both position and statistical criteria is then used to update the calibration prior to spectrum analysis. If the number of energies left in the subset is less than the number of coefficients in the fitting polynomial (order+1), no update is made.

Once established, the above set (isotope data and internal energy calibration data) is included routinely with every tape analyzed. Cards specific for processing a given tape follow this set.

1. Label Card LAB1, LAB2, (2A4) label of the tape being treated (e.g. MT27)

2. Parameter Card

NFW	NDEL	NBASE	NCUT	LWD	SFLUX
I4	I4	I4	I4	I4	I10.5

For each gamma energy an interval \pm NFW channels centered on the calculated peak position, ICH, is searched for the absolute maximum channel, IMX. The net peak area is calculated over an interval \pm NBASE channels centered on the IMX. As in the case of the energy calibration, the maximum must be within the interval $ICH \pm NFW$ to be considered correctly located. If the maximum occurs at the ends of the interval a warning is generated.

A measure of the statistical significance of each net sum over the interval $IMX \pm NBASE$, is computed and printed out. The statistical criterion is defined as $CRIT = ((N-B)/\sqrt{B})^2$ where N = total counts over the interval and B = the corresponding baseline sum. Thus CRIT is the square of the ratio of net peak counts to the standard deviation in baseline counts in the interval. This expression for CRIT can be related to the decision, detection and quantitative determination limits L_C , L_D and L_Q defined by Currie⁽²⁵⁾ in a careful discussion of signal detection thresholds. L_C is the value of that net signal which can just be detected in a single observation. L_D is the value of that true limiting mean signal which can be reliably detected in a given analytical procedure and L_Q is that level at which an individual signal is likely to be close to the limiting mean (somewhat arbitrarily taken as being within 10% with a 95% probability). "Working" expressions for the three limits are given⁽²⁵⁾ for both Gaussian and Poisson signal distributions. For the latter case

$$\begin{aligned}
 L_C &= 2.33 \sqrt{\mu_B} \\
 L_D &= 2.71 + 4.65 \sqrt{\mu_B} \\
 L_Q &= 50 (1 + \sqrt{1 + \mu_B/12.5})
 \end{aligned}$$

for "paired observations" of signal and background where μ_B = the limiting mean value of the background = σ_B^2 . For sufficiently large μ_B ,

$$\left(\frac{L_C}{\sigma_B}\right) \sim 2.33, \left(\frac{L_D}{\sigma_B}\right) \sim 4.65, \left(\frac{L_Q}{\sigma_B}\right) \sim \frac{50}{\sqrt{12.5}}$$

The approximations are correct to within 10% for $\mu_B \gg 70, 34, 1250$ counts respectively.

In the present case the net "signal" is N-B while the limiting mean background $\mu_B = B$ and $\sigma_B = \sqrt{B}$. Thus

$$\text{CRIT}_C = \left(\frac{L_C}{\sigma_B}\right)^2 \sim 6$$

$$\text{CRIT}_D = \left(\frac{L_D}{\sigma_B}\right)^2 \sim 20$$

$$\text{CRIT}_Q = \left(\frac{L_Q}{\sigma_B}\right)^2 \sim 200$$

From examples of isolated peaks having different values of CRIT, shown in fig. 14, it can be seen that there is qualitative agreement with the above results. For $\text{CRIT} \lesssim 5$ there is essentially no indication of a peak. The values of α are also measured and indicated, where

$$\alpha \equiv \frac{\text{Maximum count in the scan interval} - \bar{B}}{\sqrt{\bar{B}}}$$

where $\bar{B} = (B/\text{number of channels in the sum}) = \text{average baseline count per channel}$ in this region. Here $\alpha \sim 1$ indicating that the maximum peak count is comparable to the background fluctuations. For $\text{CRIT} \sim 10$ there is a detectable peak but little of quantitative use. The "peak" maximum is typically two standard deviations above the mean background ($\alpha \sim 2$). For $20 \lesssim \text{CRIT} \lesssim 200$ detection is reliable but fractional uncertainties in the peak area determination exceed 10%. These results are summarized below. The values entered apply with $\mu_B > 500$ counts.

Qualitative meaning of the statistical measure CRIT

CRIT	α	Comment
≤ 5	≈ 1	No detectable peak
5-20	~ 2	Detected with marginal reliability
20-200	3-10	Reliably detected but poor statistics
> 200	≥ 10	Statistics better than 10%

NDEL defines the interval for the internal energy calibration and for the titanium (320 KeV peak) flux monitor peak search. With 1KeV/channel calibration NDEL is typically taken as 5 channels.

NCUT = the number of channels out of the actual spectrum which will be treated by the program. Entering the minimum value consistent with retrieval of desired information will decrease running time. Gamma energy entered in the isotope data set with peaks in channels beyond NCUT will be deleted. (e.g. the 3083 KeV Ca^{41} peak should be analyzed. The baseline should be calculated correctly $2\ell+1$ = channels beyond this point so NCUT is taken as 3200).

LWD = the moving average half-width for the baseline calculation (BASE)
(e.g. LWD= ℓ =30)

SFLUX = the measure of reactor flux, in Ti^{320} KeV gamma counts, to which the values FK1, DF1, FK2, DF2 are referred. (e.g. 6000 counts. This number was obtained from computer analysis with NDEL=5).

3. The main control cards containing coded instructions for processing individual spectra appear next in the sequence.

ITG	NREC	NBG	ITY	NBE	NSK	NRW	NBKS	NCAL	FTR	CONV	TIME
I4			812							3F10.3	

ITG = tagword sought

NREC = 1,2,4 coded channel number 1=1024, 2=2048, etc.

NSK >0: skip NSK records

NBKS >0: backspace NBKS records

NRW >0: rewind the tape

ITG should not be entered for these operations. One instruction per card should be entered. At the end of execution of the instruction, the program expects another main control card.

NCAL <0: read energy calibration from cards in the following format:

EG	POS
2F	10.5

EG = actual gamma energy

POS = observed peak position

The order of the fit is - NCAL. Up to 100 such pairs may be entered. The end of this data set is indicated by a blank card. At the end of the calibration calculation, the program expects to read another main control card, so ITG should not be entered when NCAL<0.

NCAL>0: after reading in the spectrum with tagword ITG an internal energy calibration of order NCAL is made before analyzing.

No-operation values for the above quantities are thus zero. A totally blank card indicates the end of all data processing.

ITY = run type index defined earlier.

NBG = code describing the type of processing required for the spectrum.

		NGB				
		1	2	3	4	5
ITY	1	Sample Spectrum			Flux Monitor	Reset Flux
	2	Type of substrate			Spectrum	correction
	3	is			Standard	* Standard *
	4	POL	SOR	POR	Spectrum	Spectrum
					No Multitplier	With Multiplier

*Blank is ASH

NBE = 0 use FK1, DF1 with ITY=1,2 not used with ITY=3,4

= 1 use FK2, DF2

FTR = fraction of substrate (POL, SOR, POR or ASH for the standard) actually used.

CONV = divides the calculated weight of sample to convert to desired units
(e.g. CONV = no of cubic meters of air sampled in which case output is in $\mu\text{g}/\text{m}^3$)

If ITY=3,4 and NBG=5, that is for a standard spectrum with modified standard values CONV = the multiplier of SM for those values of FMY in the isotope data set which are left blank.

For values of ITY=1,2 NBG=4,5

and ITY=3,4 NGB=4

CONV is not used.

TIME = time in the format hours.minutes for the onset of counting either the standard or the sample. This value is needed only for ITY=3,4.

The program calculates the correct fraction in hours from this value. Since the standard and the sample cannot be counted simultaneously a correction must be made for the decay of one relative to the other. Only the relative values of time are needed in the calculation.

Sample output. Computer output listed on pages 73 to 75 results from the main control data card sequence listed on p.61. The first card with NCAL=-1 produces a linear external energy calibration prior to reading any spectra using the energy cards which follow. The associated output gives the number of iterations required to produce a fit with $\epsilon=0.0011$, the linear coefficients with the intercept = -0.207 channels in this case and slope = 1.0037 channels/KeV. The five output columns are

E C_O C_C ΔC CRIT

E = isotope gamma energy

C_O = observed peak position

C_C = peak position-calculated from the best fit

ΔC = C_O-C_C

In the case of the external calibration CRIT is undefined and taken to be zero.

At the end of the external calibration a card with NSK=8 is read and 8 records are skipped. The spectrum with tagword 9 is sought and is processed as a flux monitor spectrum. The associated output for this analysis includes a printout of the main control card image, the current energy calibration parameters and

ICH = the expected peak location (319)

IMX = the observed maximum in the scan interval (320)

"FLUX" = the net peak area of the 320 KeV Ti^{51} gamma (6970.4) with the associated Poission-statistical uncertainly (89.5)

NBE = index of the set of conversion factors to be updated (in this case FK1)

SFLUX = the net peak area on which the conversion factors entered on the isotope data cards are based.

FL = the ratio SFLUX/FLUX

The flux monitor spectrum for the short runs (400 and 1000 sec.) must be analyzed first before treating the sample spectra. In typical measurements the flux spectrum is generated after the 400 second count and before the 1000 second count. Thus in order to analyze the 400 second spectrum the computer is instructed via the next card with NBKS=2 to backspace two records. Spectrum with tagword 8 is then sought and treated as a 400 second spectrum. (ITY=1)

Here NBC=2 indicating substrate type two (SOR) was used but only a fraction equal to 0.164 of the amount entered in the isotope data cards is to be subtracted.

CONV = $0.0053 = .001 \times 5.3 \text{ m}^3$ of air were sampled by the substrate section and the output units are to be expressed in ng/m^3 while the conversion factors are in $\mu\text{g}/\text{count}$. (S is entered in mg/count so that the output units for this case is $\mu\text{g}/\text{m}^3$).

Following the listing of the control data in the output for tagword 8 are the results of analysis for each isotope present in the isotope data set within the range of NCUT. This table has the following entries

IND, ICH, IMX, NS, E, CT, DCT, W, DW, PCT, FC, CRIT

IND = the list index

ICH = expected peak channel

IMX = absolute maximum in the scan interval $ICH \pm NFW$

NS = element symbol

E = gamma energy

CT = net peak area (raw data counts)

DCT = Poission statistical uncertainty in CT

W = the corrected and converted net peak area (here in ng/m^3)

DW = the combined uncertainty in this value

PCT = the percent of substrate in the total sample weight

FC = the decay correction factor applied by the program. For ITY=1,2 FC=1.0

CRIT = the statistical criterion defined earlier.

It is assumed that the live time operating mode handles the dead time correction properly and that short-lived isotope dead time effects not correctable by live time operation are small. This is likely to be true if the overall dead time is kept low (e.g. <20%). Also it is assumed that the short-lived counting is for the same live-time period (400 and 1000 sec) as for the measurements of corresponding FK's. If not the correction must be included in the value of CONV.

W is given by

$$W = (CT \cdot FK - FTR \cdot BL) / CONV$$

where CT, FTR, and CONV are defined earlier. FK is either FK1 or FK2 and BL is the activity of the appropriate blank, POL, POR, etc. The error in W is given by

$$DW = \sqrt{\left(\frac{\partial W}{\partial FK}\right)^2 (DF)^2 + \left(\frac{\partial W}{\partial CT}\right)^2 (DCT)^2 + \left(\frac{\partial W}{\partial BL}\right)^2 (DBL)^2}$$

where DBL = DP, DS, etc.

For the first isotope, S37 (3102.4 KeV), the peak is not found within the 7 channel scan interval centered on channel 3112 but at the end channel, 3109. The net sum in the interval centered around 3109 is evaluated but the remaining calculation is deleted and a warning message printed. Inspection of the value CRIT suggests that the value is of doubtful statistical significance. This can be seen further from inspection of the original data which is presented below the isotope table. The contents of 23 channels centered on ICH are printed-out so that computer results may be readily checked by hand in those cases where any doubt about the analysis exists. The form of this table is

1. Index line 0 - 22
2. IND, IS, COUNTS (IS to IS+22) in I5 FORMAT
- .
- .
- .
- .

Thus in the case of S37 with IND=1 IS=3101, the column labeled 11 corresponding to IS+11=3112=ICH, is the expected peak position.

Upon completion of analysis of spectrum 8, 48 records are skipped and spectrum 59 is sought. This is treated as a long-irradiation standard spectrum (ITY=3, NBG=4). Comparison of the card input with the control line print out (p. 74) indicates that that time entered as 9h.15 min. has been correctly converted to 9.25 hrs. Also a new energy calibration update has been made which is not indicated in the write up, hence the new linear calibration coefficients - 0.522 and 1.002. Output for the standard spectrum has a slightly different form:

IND, ICH, IMX, NS, E, CT, DCT, FKS, DFS, PCT

where all entries have been defined previously except

FKS = calculated conversion factor ($\mu\text{g}/\text{count}$)

DFS = Poisson statistical uncertainty in CT combined with the blank uncertainty DA.

FKS = $(SM + ASH * FTR) / CT$

and DFS =

$$\sqrt{\left(\frac{\partial FKS}{\partial ASH}\right)^2 (DA)^2 + \left(\frac{\partial FKS}{\partial CT}\right)^2 (DCT)^2}$$

and it is assumed that there is no uncertainty in the value of SM or FTR.

It should be noted that while DFl is based on a series of measurements indicating the overall reproducibility of FKL, DFS is based on a single measurement. While it includes the established uncertainty in the substrate activity (DA) it is not strictly analogous to DFl.

PCT = the percent of ASH in the total activity = $100 \times \text{ASH} / (\text{SM} + \text{ASH})$

With analysis of the standard spectrum completed, one spectrum is skipped and spectrum 61 is analyzed as a sample spectrum (NBG=2, ITY=3) (p. 75). At the end of this analysis the job terminates.

Further information on all programs can be obtained from Dr. John Robbins, Great Lakes Research Division, I.S.T., North University Building, Room 1112, Telephone 764-2420.

A Sample Short-Irradiation Output from SPAN.

EXTERNAL CALIBRATION: ITERATIONS = 2 -0.206ASE 01 0.10037E 01
 511.000 511.000 0.36 -0.00
 511.000 511.000 0.36 -0.00
 3683.000 3683.000 0.13 -0.00
 846.900 846.900 -0.30 -0.00

TAPE ID: MT27 TCM= CC09 LIVE TIME= 20 SEC

CONTROL LINE: 9 4 4 1 0 C 0 0 0 0 0 0 -2.088460 1.003741

FLUX CAL.: ICH= 319 IM= 320 *FLUX*= 6970.4 89.5 NE= 1 SFLUX= 6000.0 FL= 0.86078

TAPE ID: MT27 TCM= CC08 LIVE TIME= 400 SEC

CONTROL LINE: 8 4 2 1 0 C 0 0 0 0 0 0 0.0035 0.0 -2.088460 1.003741

IND	ICH	IME	MS	E	CT	DC	W	IN	EC	FC	GMT
2	202	3109	537	3102.40	4.3	3.1	PEAK NOT WITHIN	7 CHANNEL SCAN INTERVAL			6.75
3	3092	3092	C449	3683.00	82.4	9.5	3036.112	555.376	0.5	1.0000	181.05
5	1783	1783	AL28	1778.50	463.8	99.5	1414.226	92.859	0.2	1.0000	343396.25
6	1041	1041	C066	1039.30	4112.9	92.5	3165.232	290.762	0.2	1.0000	599.22
7	319	320	T151	320.00	535.3	91.3	256.571	48.728	2.9	1.0000	30577.41
9	2762	2760	N424	2753.00	57.7	98.1	1137.360	36.200	5.5	1.0000	19.05
10	848	848	M056	846.50	6568.0	102.9	92.632	4.730	0.2	1.0000	26751.95
11	1645	1646	C138	1642.00	224.3	36.5	-51.003	227.537	112.3	1.0000	90.72

IND	IS	0	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	
1	101	0	0	1	1	0	2	1	2	1	0	1	0	0	0	0	1	0	0	0	0	0	0	1	0
2	081	0	5	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
4	172	91	75	67	112	123	128	132	197	407	958	2148	2760	2084	980	297	84	1	0	20	14	10	6	6	0
6	1870	200	181	378	377	374	190	189	224	379	440	362	283	203	180	187	186	164	167	209	164	170	183	182	182
7	368	568	577	592	560	602	632	609	588	615	600	611	643	683	695	600	637	566	617	566	571	544	535	544	544
9	3900	163	171	183	179	141	165	178	175	170	176	215	196	189	193	178	195	180	151	165	155	161	154	171	171
10	837	322	280	327	307	325	305	414	506	624	1165	2035	2450	1532	642	315	23	0	4	214	23	21	23	22	0
11	1635	76	94	121	105	63	62	52	82	96	197	126	152	120	92	96	70	87	75	67	81	75	71	72	72

A Sample Long-Irradiation Unknown Output from SPAN.

TAPE ID: WT77	TCM= CG1	LIVE TIME= 2000 SEC	CONV	TIME	DATE	Calibration Coefficients					
CONTROL LINE: 61	4 2 3 0 0 0	0.3280	0.0113	9.5000	0.55240	-1.002185					
IND	ICH	IMX	NS	E	CT	DET	V	FW	EXT	PC	CH1
1	103	103	58143	103.20	4165.4	238.4	0.352	0.009	0.0	1.0006	334.75
2	439	438	236689	438.70	766.9	187.9	90.498	2.178	0.4	1.0532	336.62
3	687	688	1187	685.70	1158.7	104.6	PEAK	180.175	0.0	1.0189	2.43
4	595	595	8882	594.30	1414.2	205.8	27.100	1.2565	0.0	1.0128	1879.70
5	595	595	8882	594.30	1414.2	205.8	27.100	1.2565	0.0	1.0128	1879.70
6	520	520	8882	519.00	8013.9	185.3	28.735	1.532	6.0	1.0128	1879.70
7	1046	1045	8882	1045.90	2416.2	151.0	20.455	2.784	5.8	1.0158	1540.98
8	1477	1478	8882	1474.70	512.7	71.8	27.620	3.498	6.4	1.0128	334.44
9	1046	1045	8882	1045.90	2416.2	151.0	20.455	2.784	5.8	1.0158	1540.98
10	1206	1207	6472	1206.40	15.6	75.7	5.880	6.375	0.0	1.0350	2.85
11	985	984	51252K	983.50	806.0	130.5	0.0645	0.013	0.0	1.0491	659.52
12	985	984	51252K	983.50	806.0	130.5	0.0645	0.013	0.0	1.0491	659.52
13	598	598	476	597.00	13228.4	203.5	PEAK	NOT WITHIN	7 CHANNEL SCAN INTERVAL		1242.52
14	598	598	476	597.00	13228.4	203.5	PEAK	NOT WITHIN	7 CHANNEL SCAN INTERVAL		1242.52
15	1218	1216	4576	1215.80	421.3	114.8	11.528	4.412	0.0	1.0173	27.62
16	1524	1529	4576	1524.80	3243.7	109.9	24.988	2.863	0.0	1.0173	10.04
17	1524	1529	4576	1524.80	3243.7	109.9	24.988	2.863	0.0	1.0173	10.04
18	1558	1559	45140	1555.10	4767.7	76.7	68.640	0.262	0.6	1.0113	177.62
19	359	358	58142	358.60	3441.4	247.6	5.982	1.175	0.2	1.0113	409.49
20	359	358	58142	358.60	3441.4	247.6	5.982	1.175	0.2	1.0113	409.49
21	493	493	58122	492.50	2798.7	152.6	PEAK	NOT WITHIN	7 CHANNEL SCAN INTERVAL		705.10

- 1) R.C. Koch, "Activation Analysis Handbook", Chemistry Dept. Nucl. Sci. and Eng. Corp., Pittsburgh 30, Pa., (Dec. 1958).
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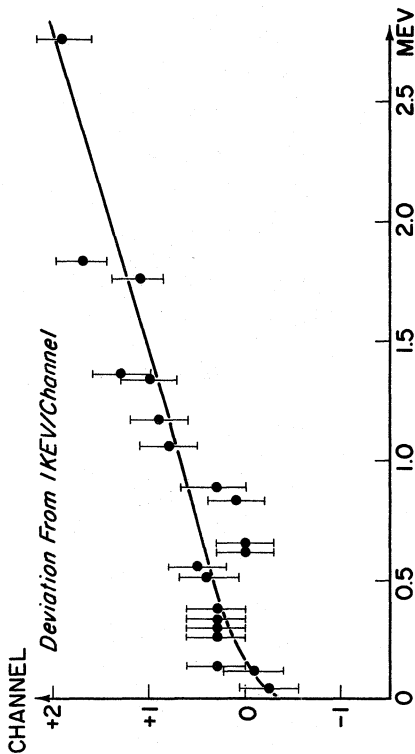


Fig. 1

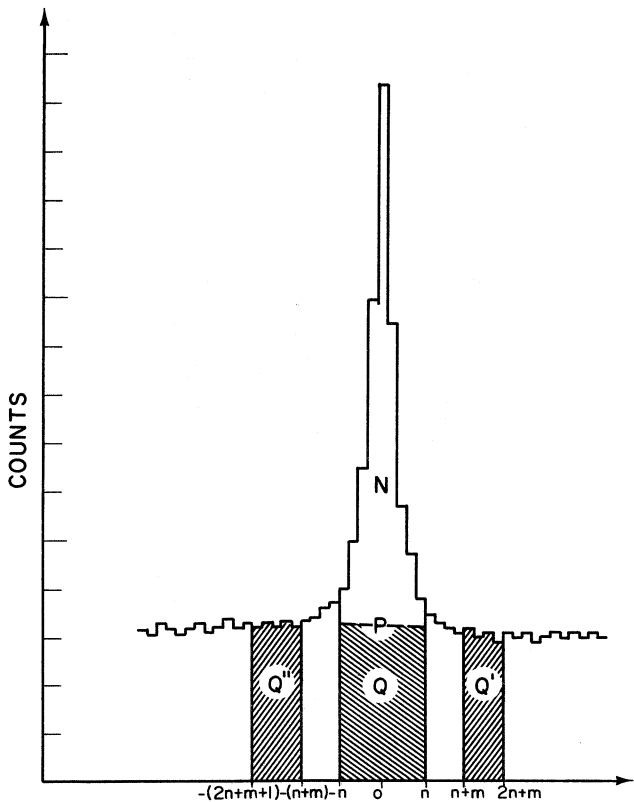
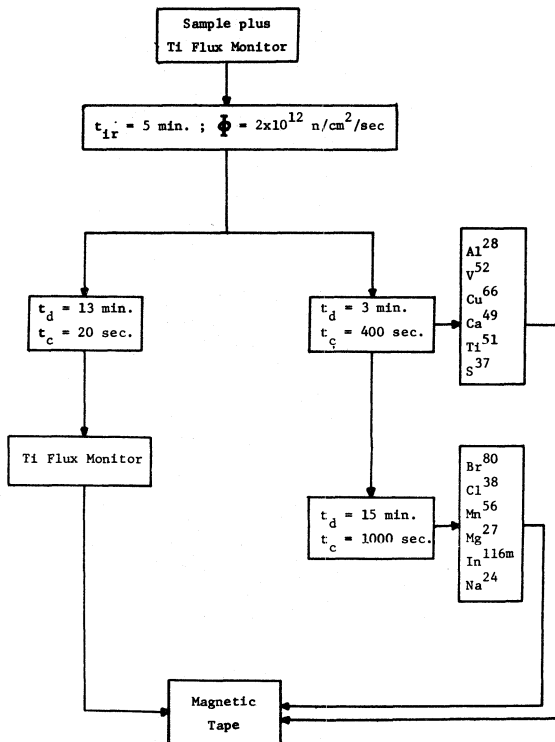


Fig. 2

Fig. 3

Irradiation and Counting Scheme for Short-lived Isotopes.



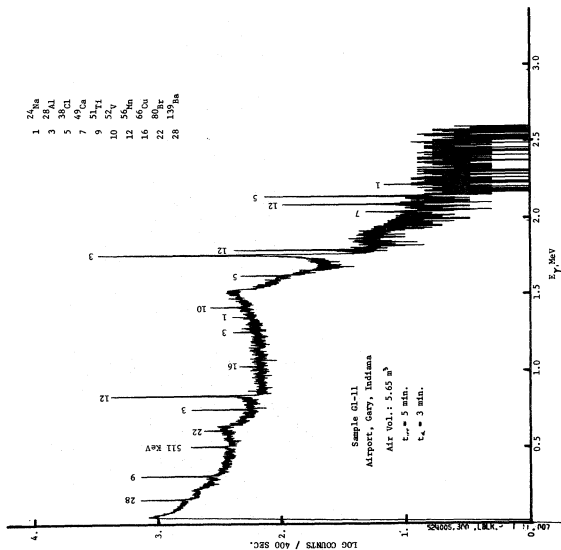


Fig. 4

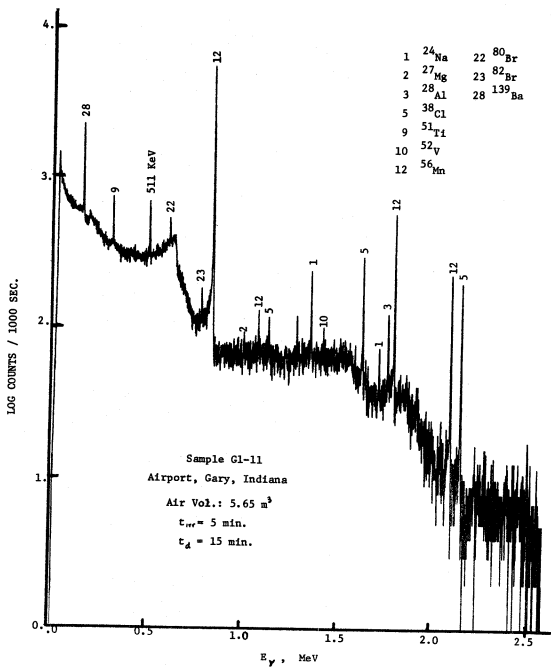
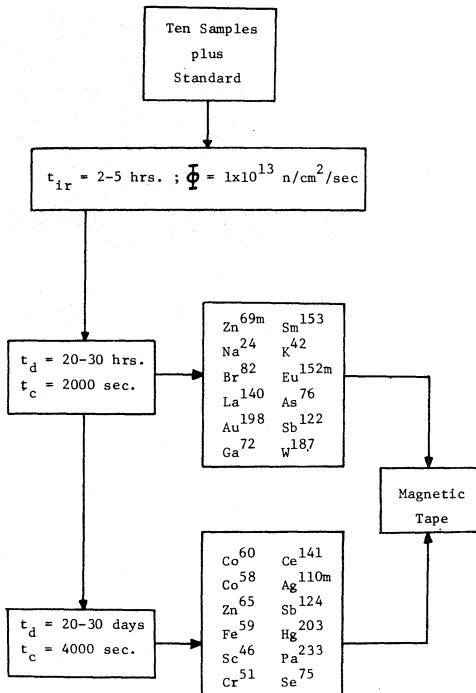


Fig. 5

Fig. 6

Irradiation and Counting Scheme for Long-lived Isotopes.



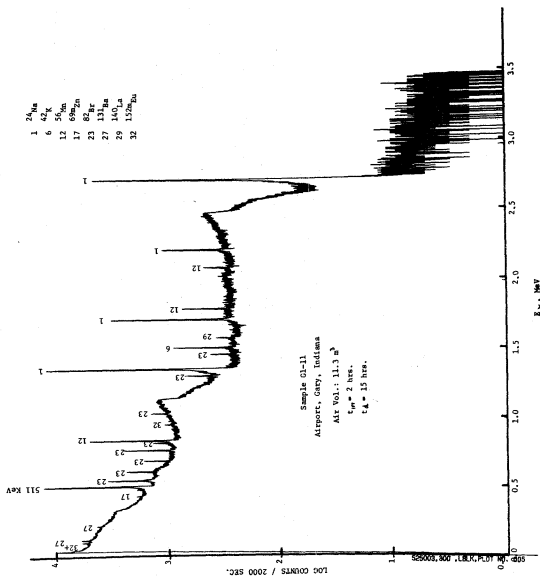


Fig. 7

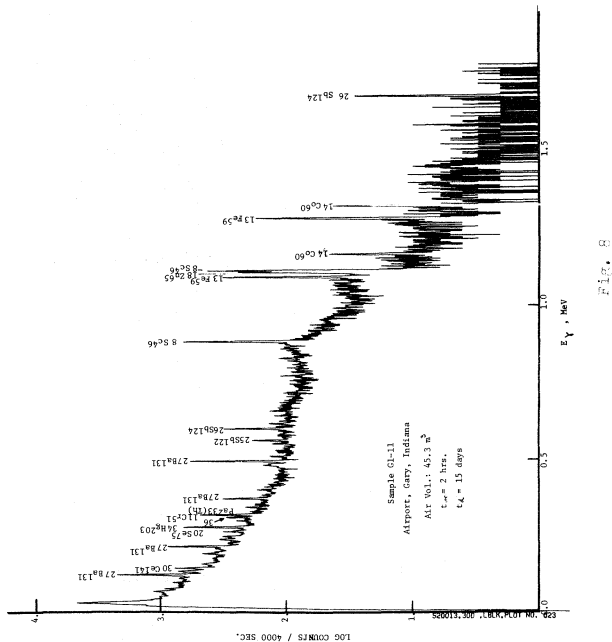


Fig. 9

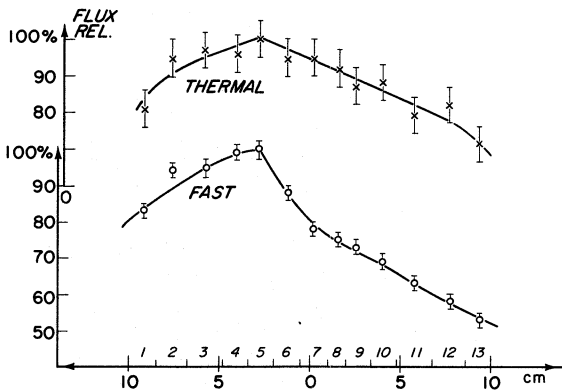
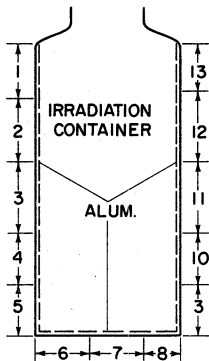
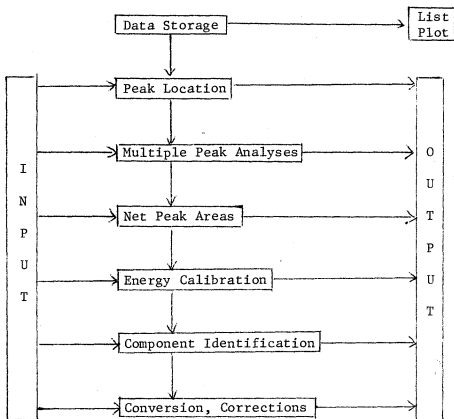


Fig. 10. A General Data Reduction Schematic Diagram



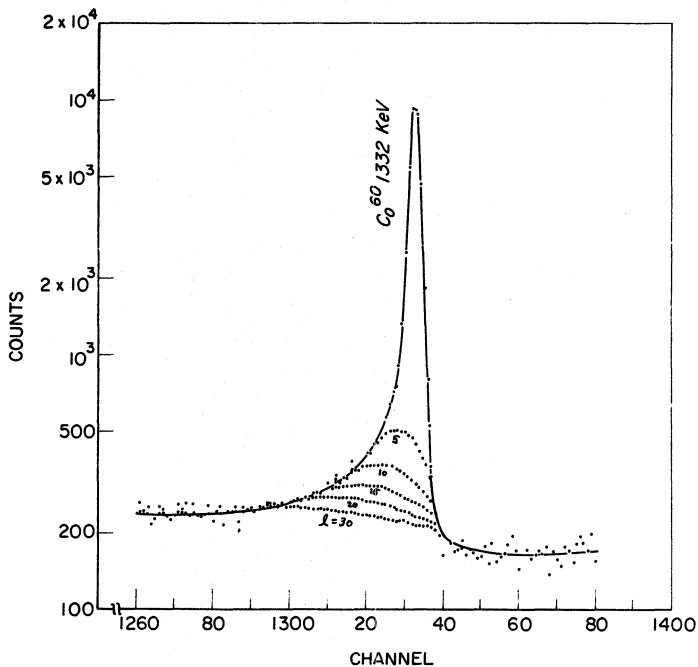


Fig. 11 Dependence of the calculated baseline on the moving-average half width, l .

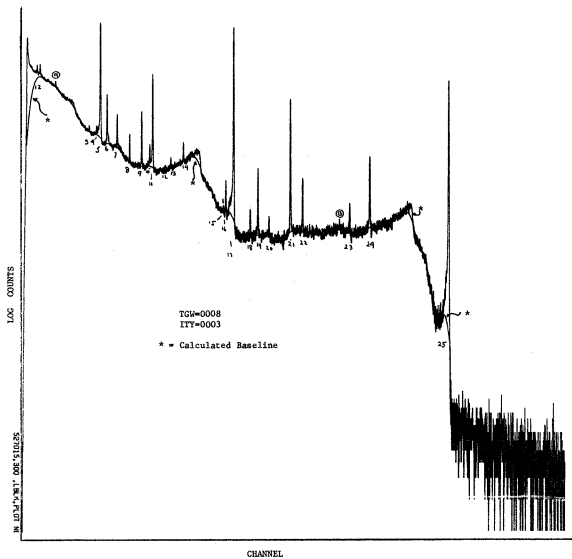


Fig. 12. Gamma spectrum showing the calculated baseline and computer-located peaks.

An Abbreviated Flow Diagram for SPAN .

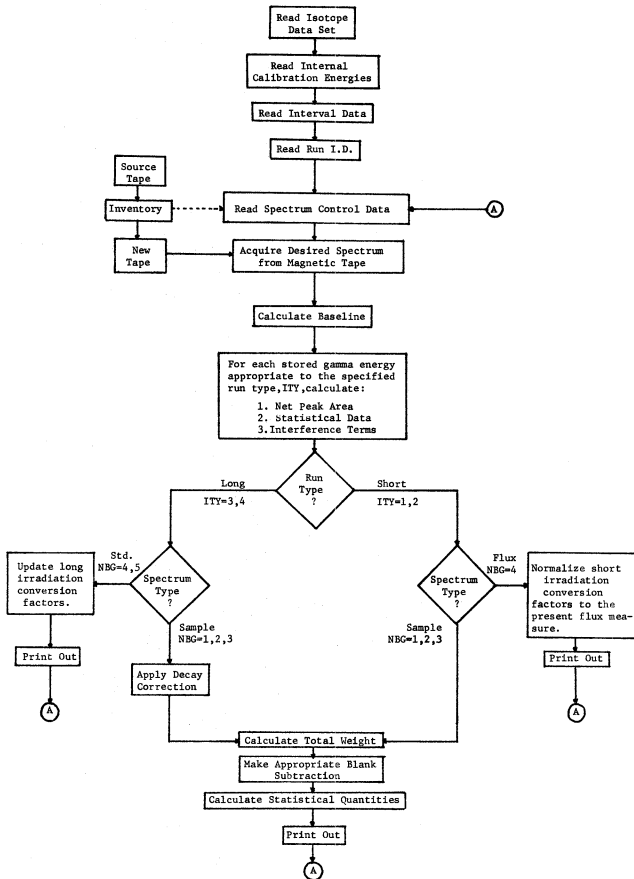


Fig. 13.

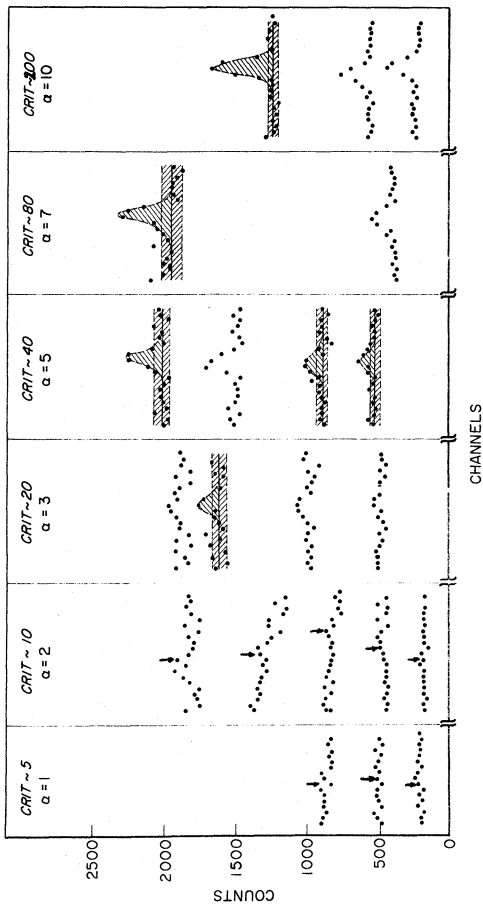


Fig. 14 Sample spectrum peaks versus the statistical criterion, crit. (Arrows indicate expected peak positions.)

APPENDIX

UL MEMORY REQUIREMENTS 078C68, BYTES.

The RDLST and BASE Programs :

COMPILER	DATE	05-11-72	17:26.31	PAGE 0701	N. IV. S. COMPILER	BASE	03-28-70	10:11.26	PAGE 0001
C MAIN HEAD 1351 PRECOMP IMPLICIT PRECISION REAL*80 COMMON PRECISION IND*8000 CALL RDLST 100 DIMENSION IND(10000) CALL RDLST(IND,1) 2 STOP 3 CONTINUE CALL RDLST(IND,1) CALL RDLST(IND,1) 200 DIMENSION IND(10000) CALL RDLST(IND,1) IF (IND(1)) GO TO 200 4 CONTINUE CALL RDLST(IND,1) 111 CONTINUE CALL RDLST(IND,1) 112 CONTINUE CALL RDLST(IND,1) CALL R									

A Simple Main Plotting Program :

FORTRAN IV G COMPILER		MAIN	10-24-69	15:04.22	PAGE 01
0001		DOUBLE PRECISION RS(4096)			
0002		DIMENSION SR(4096),CH(4096)			
0003		DIMENSION IR(4095)			
0004		FTY=2.0			
0005		FTX=.0025			
0006		XX=66.			
0007		CALL PLTXMX(XMX)			
0008		XST=11.0			
0009	999	CONTINUE			
0010		READ(5,420) NREC,ITG,NCUT,NSK,NCR			
0011		CALL SKIP(0,NSK,4)			
0012	420	FORMAT(514)			
0013		IF(NREC)805,805,717			
0014	717	CONTINUE			
0015		NR=NREC-1			
0016	478	CONTINUE			
0017		CALL RDTAPE(NR,13,1T,IR)			
0018		IF(MVL(13)-ITG)478,799,805			
0019	805	STOP			
0020	799	CONTINUE			
0021		XST=XST+11.0			
0022		XND=XST+10.0			
0023		CALL PENUP(XST,0.5)			
0024		CALL PENDN(XST,0.5)			
0025		CALL PENDN(XND,0.5)			
0026		CALL PENUP(XST,0.5)			
0027		CALL PENDN(XST,0.0)			
0028		DO 1 J=1, NCUT			
0029		SR(J)=IR(J)			
0030		IF (SR(J)) 2,2,3			
0031	2	CONTINUE			
0032		SR(J)=0.5			
0033		GO TO 987			
0034	3	SR(J)=0.5+FTY*ALOG10(SR(J))			
0035	987	CONTINUE			
0036		U=J			
0037	1	CH(J)=U+FTX+XST			
0038		CALL PLINE(CH(1),SR(1),NCUT,1,0,0,1)			
0039		IF(INCR)998,999,998			
0040	998	CONTINUE			
0041		CALL BASE(30,1,NCUT,IR,85)			
0042		DO 51 J=1, NCUT			
0043		T=BS(J)			
0044		IF(T-1.0)52,52,53			
0045	52	T=1.0			
0046	53	CONTINUE			
0047	51	SR(J)=0.5+FTY*ALOG10(T)			
0048		CALL PLINE(CH(1),SR(1),NCUT,1,0,0,1)			
0049		GO TO 999			
0050		END			

[illegible]

Non-linear Least Squares Routines :

```

IV C COMPILER      ALLS      12-31-69      11:26:11      PAGE 0001
SUBROUTINE NLLS(X,Y,N,D,G,P,SE,VAR,ERR,EPS,NIT,ND,NP,K)
C NCL=LINEAR LEAST SQUARES FITTING ROUTINE
C (X,Y PAIRS MUST BE DESCRIBED BY A CONTINUOUS AND DIFFERENTIABLE FUNCTION
C FIX,G(J)) WHERE G(J)'S ARE ADJUSTABLE PARAMETERS. NLLS STARTS WITH GUESSES
C FOR G(J) AND HELDONS *BEST FIT* VALUES FOR THEM WHICH MINIMIZE THE
C SUM OF THE SQUARES OF THE DIFFERENCES BETWEEN Y AND F.
C ND=NO. OF CATAPOINTS X(J),Y(J)
C W=STATISTICAL WEIGHT ASSOCIATED WITH Y(J). 1/STANDARD DEVIATION**2
C G(J)=GUESS PARAMETERS INITIALLY. *BEST FIT* PARAMS. UPON RETURN FROM NLLS
C P=COLUMN PARAMETER COMMUNICATING WITH ARG
C SE(J)=STANDARD ERROR IN PARAMETER G(J)
C VAR=VARIANCE OF FIT
C ERR=0 UNLESS DET OF INVERSE MATRIX =0.0, THEN ERR=1
C NIT=NO. OF ITERATIONS ALLOWED
C K=ACTUAL NUMBER PERFORMED
C EPS=ALLOWED FRACTIONAL ERROR IN EVERY G(J) TO TERMINATE ITERATION
C NP=NO. OF PARAMETERS (J)
C DOUBLE PRECISION T(20,20)
C DIMENSION X(100),Y(100),W(100),G(20),SE(20),R(20),D(20),P(10)
C DIMENSION I(20,20)
C DIMENSION IP(40)
C DIMENSION CC(20)
C ERR=0
C DO 1 I=1,ND
C IF(W(I))12,2,1
C 2 NIT=NIT+1
C 1 CONTINUE
C K=0
C 3 IF(I-NIT)4,11,11
C 4 K=K+1
C DO 5 J=1,NP
C R(J)=0.0
C DO 5 I=1,NP
C 5 AT(I,J)=0.0
C DO 6 I=1,ND
C CALL ARGIV(X(I),G,G,P)
C OI=X(I)-Y(I)+W(I)
C DO 6 J=1,NP
C R(J)=R(J)+O(I)*D(I,J)*W(I)
C DO 6 L=1,NP
C AT(L,J)=AT(L,J)+O(I)*O(I)*G(I,J)*W(I)
C 6 T(L,J)=AT(L,J)
C DO 7 J=1,NP
C DO 7 L=1,NP
C 7 T(L,J)=T(L,J)
C CALL INVNP(20,T,IP,20,T)
C DO 8 J=1,NP
C CC(J)=0.0
C DO 8 I=1,NP
C OUM=1./T(I,J)
C 8 CC(J)=CC(J)+O(I)*O(I)*W(I)
C DO 14 J=1,NP
C IF(ABS(T(I,J)-1.0CC-30))15,14,14
C 15 ERR=1.0
C 14 CONTINUE
C IF(ERR)16,16,17
C 17 RETURN
C 16 CONTINUE
C KCHK=0
C DO 9 J=1,NP
C IF(G(J))18,8,914
C 914 CONTINUE
C IF(ABS(CC(J)/G(J))-EPS)8,8,10
C 10 KCHK=1
C R(G(J))=G(J)+CC(J)
C IF(KCHK)11,11,13
C 11 VAR=0.0
C DO 12 I=1,ND
C CALL ARGIV(X(I),G,G,P)
C 12 VAR=VAR+(I-IP(I))-V**2
C RG=ND-NP
C IF(RG)1001,1001,1002
C 1001 VAR=0.0
C DO 10 1003
C 1002 CONTINUE
C VAR=SORT(VAR/PGT)
C 1003 CONTINUE
C DO 13 J=1,NP
C OUM=DABS(T(J,J))
C 13 SE(J)=VAR* SORT(OUM)
C RETURN
C END

```

MEMORY REQUIREMENTS 00014 BYTES

[illegible]

SPAN List Continued :

IN IV G COMPILER	MAIN	23-28-70	1128-04	PAGE 0005 IN IV G COMPILER	MAIN	03-28-70	1128-04	PAGE 0007
710	CONTINUE							
711	CALL IMTC(MP,18,18)							
712	END							
713	IF (MTC(1,1) - 1) GO TO 714							
714	CONTINUE							
715	CALL IMTC(MP,18,18)							
716	END							
717	IF (MTC(1,1) - 1) GO TO 718							
718	CONTINUE							
719	CALL IMTC(MP,18,18)							
720	END							
721	IF (MTC(1,1) - 1) GO TO 722							
722	CONTINUE							
723	CALL IMTC(MP,18,18)							
724	END							
725	IF (MTC(1,1) - 1) GO TO 726							
726	CONTINUE							
727	CALL IMTC(MP,18,18)							
728	END							
729	IF (MTC(1,1) - 1) GO TO 730							
730	CONTINUE							
731	CALL IMTC(MP,18,18)							
732	END							
733	IF (MTC(1,1) - 1) GO TO 734							
734	CONTINUE							
735	CALL IMTC(MP,18,18)							
736	END							
737	IF (MTC(1,1) - 1) GO TO 738							
738	CONTINUE							
739	CALL IMTC(MP,18,18)							
740	END							
741	IF (MTC(1,1) - 1) GO TO 742							
742	CONTINUE							
743	CALL IMTC(MP,18,18)							
744	END							
745	IF (MTC(1,1) - 1) GO TO 746							
746	CONTINUE							
747	CALL IMTC(MP,18,18)							
748	END							
749	IF (MTC(1,1) - 1) GO TO 750							
750	CONTINUE							
751	CALL IMTC(MP,18,18)							
752	END							
753	IF (MTC(1,1) - 1) GO TO 754							
754	CONTINUE							
755	CALL IMTC(MP,18,18)							
756	END							
757	IF (MTC(1,1) - 1) GO TO 758							
758	CONTINUE							
759	CALL IMTC(MP,18,18)							
760	END							
761	IF (MTC(1,1) - 1) GO TO 762							
762	CONTINUE							
763	CALL IMTC(MP,18,18)							
764	END							
765	IF (MTC(1,1) - 1) GO TO 766							
766	CONTINUE							
767	CALL IMTC(MP,18,18)							
768	END							
769	IF (MTC(1,1) - 1) GO TO 770							
770	CONTINUE							
771	CALL IMTC(MP,18,18)							
772	END							
773	IF (MTC(1,1) - 1) GO TO 774							
774	CONTINUE							
775	CALL IMTC(MP,18,18)							
776	END							
777	IF (MTC(1,1) - 1) GO TO 778							
778	CONTINUE							
779	CALL IMTC(MP,18,18)							
780	END							
781	IF (MTC(1,1) - 1) GO TO 782							
782	CONTINUE							
783	CALL IMTC(MP,18,18)							
784	END							
785	IF (MTC(1,1) - 1) GO TO 786							
786	CONTINUE							
787	CALL IMTC(MP,18,18)							
788	END							
789	IF (MTC(1,1) - 1) GO TO 790							
790	CONTINUE							
791	CALL IMTC(MP,18,18)							
792	END							
793	IF (MTC(1,1) - 1) GO TO 794							
794	CONTINUE							
795	CALL IMTC(MP,18,18)							
796	END							
797	IF (MTC(1,1) - 1) GO TO 798							
798	CONTINUE							
799	CALL IMTC(MP,18,18)							
800	END							
801	IF (MTC(1,1) - 1) GO TO 802							
802	CONTINUE							
803	CALL IMTC(MP,18,18)							
804	END							
805	IF (MTC(1,1) - 1) GO TO 806							
806	CONTINUE							
807	CALL IMTC(MP,18,18)							
808	END							
809	IF (MTC(1,1) - 1) GO TO 810							
810	CONTINUE							
811	CALL IMTC(MP,18,18)							
812	END							
813	IF (MTC(1,1) - 1) GO TO 814							
814	CONTINUE							
815	CALL IMTC(MP,18,18)							
816	END							
817	IF (MTC(1,1) - 1) GO TO 818							
818	CONTINUE							
819	CALL IMTC(MP,18,18)							
820	END							
821	IF (MTC(1,1) - 1) GO TO 822							
822	CONTINUE							
823	CALL IMTC(MP,18,18)							
824	END							
825	IF (MTC(1,1) - 1) GO TO 826							
826	CONTINUE							
827	CALL IMTC(MP,18,18)							
828	END							
829	IF (MTC(1,1) - 1) GO TO 830							
830	CONTINUE							
831	CALL IMTC(MP,18,18)							
832	END							
833	IF (MTC(1,1) - 1) GO TO 834							
834	CONTINUE							
835	CALL IMTC(MP,18,18)							
836	END							
837	IF (MTC(1,1) - 1) GO TO 838							
838	CONTINUE							
839	CALL IMTC(MP,18,18)							
840	END							
841	IF (MTC(1,1) - 1) GO TO 842							
842	CONTINUE							
843	CALL IMTC(MP,18,18)							
844	END							
845	IF (MTC(1,1) - 1) GO TO 846							
846	CONTINUE							
847	CALL IMTC(MP,18,18)							
848	END							
849	IF (MTC(1,1) - 1) GO TO 850							
850	CONTINUE							
851	CALL IMTC(MP,18,18)							
852	END							
853	IF (MTC(1,1) - 1) GO TO 854							
854	CONTINUE							
855	CALL IMTC(MP,18,18)							
856	END							
857	IF (MTC(1,1) - 1) GO TO 858							
858	CONTINUE							
859	CALL IMTC(MP,18,18)							
860	END							
861	IF (MTC(1,1) - 1) GO TO 862							
862	CONTINUE							
863	CALL IMTC(MP,18,18)							
864	END							
865	IF (MTC(1,1) - 1) GO TO 866							
866	CONTINUE							
867	CALL IMTC(MP,18,18)							
868	END							
869	IF (MTC(1,1) - 1) GO TO 870							
870	CONTINUE							
871	CALL IMTC(MP,18,18)							
872	END							
873	IF (MTC(1,1) - 1) GO TO 874							
874	CONTINUE							
875	CALL IMTC(MP,18,18)							
876	END							
877	IF (MTC(1,1) - 1) GO TO 878							
878	CONTINUE							
879	CALL IMTC(MP,18,18)							
880	END							
881	IF (MTC(1,1) - 1) GO TO 882							
882	CONTINUE							
883	CALL IMTC(MP,18,18)							
884	END							
885	IF (MTC(1,1) - 1) GO TO 886							
886	CONTINUE							
887	CALL IMTC(MP,18,18)							
888	END							
889	IF (MTC(1,1) - 1) GO TO 890							
890	CONTINUE							
891	CALL IMTC(MP,18,18)							
892	END							
893	IF (MTC(1,1) - 1) GO TO 894							
894	CONTINUE							
895	CALL IMTC(MP,18,18)							
896	END							
897	IF (MTC(1,1) - 1) GO TO 898							
898	CONTINUE							
899	CALL IMTC(MP,18,18)							
900	END							

